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Material Resources, Energy, and Nutrient Recovery from Waste: Are Waste Refineries the Solution for the Future?

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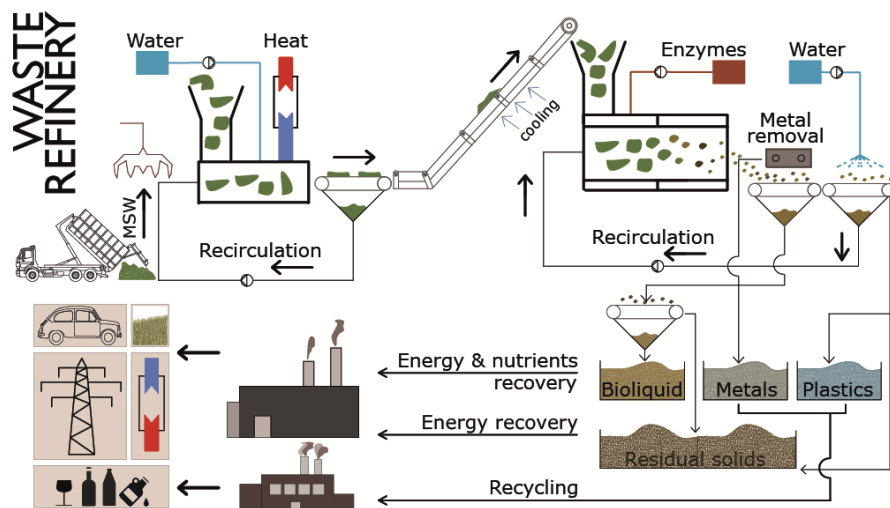
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Abstract

Waste refineries focusing on multiple outputs of material resources, energy carriers, and nutrients may potentially provide more sustainable utilization of waste resources than traditional waste technologies. This consequential life cycle assessment (LCA) evaluated the environmental performance of a Danish waste refinery solution against state-of-the-art waste technology alternatives (incineration, mechanical-biological treatment (MBT), and landfilling). In total 252 scenarios were evaluated, including effects from source-segregation, waste composition, and energy conversion pathway efficiencies. Overall, the waste refinery provided global warming (GW) savings comparable with efficient incineration, MBT, and bioreactor landfilling technologies. The main environmental benefits from waste refining were a potential for improved phosphorous recovery (about 85%) and increased electricity production (by 15-40% compared with incineration); albeit at the potential expense of additional toxic emissions to soil. Society's need for the outputs from waste, i.e. energy products (electricity vs. transport fuels) and resources (e.g., phosphorous), and the available waste composition were found decisive for the selection of future technologies. Based on the results, it is recommended that a narrow focus on GW aspects should be avoided as most waste technologies may allow comparable performance. Rather, other environmental aspects such as resource recovery and toxic emissions should receive attention in the future.



1. Introduction

Security of supply of energy and material resources may likely become an increasingly important aspect of national decision making in the future. Biomass resources for energy and chemical feedstock may be limited by constraints related to land-use and associated environmental impacts (1-4). Consequently, utilization of waste materials may become more important. Waste generation in society is related to consumption patterns, economic activity, industrial and technological level, etc. When waste has been generated, it represents a cost and a potential environmental load to society, but waste also represents a valuable source of resources that can be utilized for production of energy and materials, and a source of nutrients. With increased scarcity of resources in the future, the ability of society to recover and intelligently manage these resources becomes an important competitive advantage in a global economy (5, 6).

Traditional waste management solutions (e.g., landfilling, thermal treatment, biogas production) generally focus on providing one or two useful types of outputs (e.g., energy or recyclable materials) while at the same time minimizing the pollution associated with the waste technology. Following the concepts of biorefineries (e.g. (7-11)), new types of waste technologies have emerged promising more integrated approaches to waste management by focusing on multiple outputs addressing the need in society for securing both material resources, energy, and nutrients from the waste (12). These waste refineries potentially allow more flexible production of energy carriers from waste (gaseous, liquid, and solid fuels) while at the same time ensuring recovery of recyclable materials and nutrient resources. While the integrated approach to waste management offered by waste refinery technologies appear promising, very few studies have addressed the environmental performance of the technologies in comparison with more traditional solutions.

A Danish waste refinery technology (12), currently demonstrated in pilot-scale, separates incoming mixed household waste in two fractions: i) a bioliquid derived from enzymatic liquefaction of degradable materials such as organic waste, paper, and cardboard, and ii) a remaining solid and non-degradable fraction. Metals and plastic materials can be separated from the solid fraction for downstream recycling. The bioliquid can be digested to produce biogas or transport fuels. The remaining solids can be upgraded to Solid Recovered Fuel (SRF) for efficient combustion at power plants. Nutrients in residuals from anaerobic digestion can be further utilized when applied on agricultural land. An important potential benefit for society is

the versatility and storability of the produced energy carriers (e.g., for natural gas engines, gas turbines, boilers, and upgrading for use as transport fuel, etc.). This may allow improved environmental and economic benefits from waste, and may better match the needs in future energy systems with high shares of intermittent energy sources such as wind energy and photovoltaic (3, 13-17).

Although elaborate source-segregation of waste is an alternative to waste refineries, and various segregation techniques exist today (18, 19), the outcome may not always match the expectations. For example, source-segregated organic household waste may still contain impurities and need further pre-treatment prior to biological conversion. This may lead to considerable losses of mass, energy, and nutrients as recently documented (20). Mechanical-biological treatment (MBT) of waste addresses a similar problem as the waste refineries by combining a range of mechanical operations for separating the organic fraction from the incoming mixed waste (21-27). A share of the remaining materials is recovered for recycling while the residuals are typically utilized as SRF for energy production. The recovered organic fraction is generally biologically treated by composting or anaerobic digestion (21). A disadvantage of MBT technologies is that the stabilized organic material (compost) is often contaminated by impurities preventing further recycling of nutrients on land (22). Additionally, a low quality of the separated organic material may reduce the biogas yield during subsequent anaerobic digestion thereby lowering the overall energy efficiency of the process (21, 22, 24, 25, 27). By involving enzymes for separation of organics, the waste refinery potentially provides a more efficient solution for recovery of nutrients and energy resources in waste, at the same time allowing separation of recyclable materials to be used as raw materials in industry.

The goal of this study is to quantify the environmental impacts and energy performance associated with waste refinery solutions for 1 tonne of municipal solid waste in comparison with state-of-the-art waste management technologies (incineration, landfilling, and MBT). The study further assesses the importance of source-segregation of organic waste, waste composition, and efficiencies of involved energy conversion technologies for the overall environmental performance and for the ranking between technologies.

2. Methodology

Life cycle assessment

Scope and functional unit

The environmental assessment followed the principles of consequential life cycle assessment (28, 29). The functional unit was "treatment of 1 tonne (1 t = 1 Mg, wet weight) of municipal solid waste (MSW)". The geographical scope was Europe. The waste composition was based on recent studies investigating European waste (23, 30). The chemical composition of individual waste material fractions was based on (30). The waste treatment technologies were modelled as state-of-the-art technologies as they will likely be established in coming years: energy efficiency, emissions, and resource consumptions were extrapolated based on recent trends and expected future performances for the period 2015-2030 (for details see supporting information, SI).

All environmental impacts (resource consumption, emissions to air, soil, and water) were accounted for a 100-year time horizon. Given the functional unit of 1 tonne of waste, the activities related to the "production of waste" were excluded (since it was identical in all scenarios). This approach is sometime described as the "zero burden approach" and follows common practice within waste LCA (31). Downstream utilization of recovered energy and recyclables were credited in the LCA scenarios by system expansion into the associated energy and industrial systems (avoiding energy and virgin material production). The system boundaries included the household, starting from waste collection, transportation, treatment, disposal of residues (e.g., incineration ashes, stabilized organic material, etc.), recycling of materials, and application on land of aerobically stabilized digestate (compost) from anaerobic digestion. The environmental impacts associated with facility construction and demolition were not included due to lack of consistent data. While it should be emphasized that a full sustainability assessment should include also social and economic aspects, this was beyond the scope of the study.

Impact assessment

The following environmental impacts were included in the assessment: global warming (GW), acidification (AC), aquatic eutrophication - nitrogen (EP (N)), freshwater ecotoxicity (ETw), and human toxicity (HT). The impact assessment followed recent recommendations for best practice within LCA (32). The following assessment methods were used: IPCC 2007 for GW (33), accumulated exceedance for AC (34), EUTREND for EP (N) (35), and Usetox for ETw and HT (36). For AC, Danish regionalized characterization factors were used for SO₂, NO_x and NH₃.

These factors roughly corresponded to EU27 average values for country-specific characterization factors (34). For EP only N-eutrophication was considered, as N was the limiting nutrient in Danish aquatic ecosystems (no significant P leaching). Additionally, the resource category “phosphorous resource saving” (Pres) based on the Impact 2002+ method (37) was included in order to reflect consequences associated with use-on-land of compost produced from biological treatment.

LCA scenario modeling and system boundaries

Two different sets of waste (*a* and *b*, see section 2.2.1) as generated by the source were considered to address the intrinsic variability of MSW. Three levels of organic waste source-segregation efficiencies (0, I and II, see section 2.2.2) and seven different treatment strategies for the residual waste (i.e., waste remaining after source-segregation of recyclables) were addressed, see section “Treatment of the residual waste” for details on the latter. This resulted in 42 “baseline” scenarios (BS), see Figure 1. Each of the 42 scenarios was evaluated against six sensitivity settings (see Figure 1), in total providing 252 scenarios. For details regarding material flows, please refer to Figures S1-S14 and Table S9 in SI.

Waste composition

Table S1 summarizes the composition of the two municipal solid waste types: (*a*) Danish composition (30) and (*b*) Spanish composition (23). The principal differences between the two datasets were: i) the share (% of the total) of organic waste: in *a*, this was about 34%, while in *b* approximately 59%, and ii) the share of paper and cardboard materials which was 30% in *a* and 14% in *b*. The two waste types represented typical variations encountered in Europe and Worldwide (38-41).

Source-segregation of recyclables and organic waste

All 42 scenarios were modeled assuming household source-segregation of selected recyclable materials: I) ferrous metals, II) aluminum, III) plastic (hard plastic, i.e. high-density polyethylene, HDPE, and polypropylene, HDPP, plastic bottles, e.g., polyethylene, PET, and soft plastic such as foils, plastic bags, etc.), IV) paper and cardboard (dirty paper and dirty cardboard excluded), and V) glass. Typical source-segregation efficiencies were assumed, i.e., 75% for

ferrous metals and aluminum, 50% for plastic, 80% for paper and cardboard, and 75% for glass. These values represented average segregation efficiencies for a range of collection systems from full-service (door to door) to joint collection points (e.g., in apartments) as illustrated in (19). The segregated recyclables were sent to remanufacturing facilities for further recycling.

Three alternatives were investigated for the organic waste (i.e., vegetable and animal food waste, kitchen tissues, and wood-like materials): a scenario with no source-segregation (*O*), i.e., all organic waste ended up in the residual waste, a "realistic" scenario with 70% source-segregation efficiency (*I*), and an "extreme" scenario with 100% efficiency (*II*). Note that scenario *I* with 70% efficiency corresponded to the middle of the range provided in (19). Only vegetable and animal food waste, kitchen tissues, and wood-like materials were considered as part of the organic waste for source-segregation to minimize contamination. As source-segregation was not 100% efficient for all materials, some recyclables and organics were still present in the residual waste (rMSW). See Table S1 for details concerning composition.

Source-segregated organic waste was pre-treated for removal of un-wanted objects (e.g., plastic, large wood pieces, etc.) prior to anaerobic digestion for biogas production (and combined-heat-and-power (CHP) generation). Digestate from the process was subsequently aerobically stabilized (composted) and dried to achieve final compost quality. The compost was used as fertilizer avoiding mineral N, P, and K fertilizer production and use (calcium ammonium nitrate, diammonium phosphate, and potassium chloride, respectively (42, 43)). Substitution was based on the content of N, P, and K in the compost. See SI for further details.

Treatment of the residual waste

Seven different treatments for rMSW were considered (see Figure 1): i) waste refinery with anaerobic digestion of bioliquid and biogas combustion in natural gas engines for electricity and district heating (CHP) production (*WR GE*), ii) waste refinery with anaerobic digestion of bioliquid and upgrading of the biogas to transport fuel quality (*WR TF*), iii) incineration with CHP production (*INC*), iv) mechanical-biological treatment (MBT) with anaerobic digestion for CHP production and post-composting of the digested material (*MBT AC*), v) mechanical-biological treatment with direct composting of the organic material (*MBT DC*), vi) bioreactor landfilling with electricity production from the collected biogas (*BLF*), vii) conventional landfilling with biogas flaring (*CLF*). In waste refinery scenarios (*WR GE*, *WR TF*), compost

produced from digestate after anaerobic digestion of the bioliquid was assumed applied on land following the approach used for source-segregated organic waste. Instead, in the case of MBT, the compost (stabilized organic matter) was assumed landfilled according to current practice (22, 27).

Energy system

Electricity, heat, and transport fuel (produced from waste incineration or biogas conversion) were assumed to substitute marginal peak-load electricity, district heating, and transport fuel production following the principles of consequential LCA (28, 29). For fossil fuels used for electricity and district heating production, coal and natural gas represented two ends of the interval with respect to CO₂ emissions per fuel energy unit. At a European level, these were the most likely fossil fuels to respond to changes in electricity production (see SI). For the baseline scenarios, coal was assumed as marginal fuel for electricity. This assumption was tested in the sensitivity analysis by alternatively substituting with natural gas. For district heating, three alternatives representing Western/Southern, Eastern, and Northern European heat markets were addressed (see SI). The substituted heating technologies were: 1) natural gas boilers (Western/Southern Europe, baseline assumption), 2) coal boilers (Eastern Europe, sensitivity analysis S2), and 3) coal-fired CHP plants (Northern Europe, sensitivity analysis S3). For transport fuels, gasoline was assumed marginal in the baseline scenarios. This assumption was tested in the sensitivity analysis S5 by substituting diesel fuel.

LCI of waste technologies

The waste refinery process was based on a pilot-scale facility in Copenhagen, Denmark. The waste refinery produced two main outputs from the incoming mixed MSW: i) a bioliquid (i.e., slurry composed of enzymatically liquefied organic, paper, and cardboard) and ii) a solid fraction (i.e., non-degradable waste materials).

The refinery process comprised two reactors: in the first reactor waste was heated by injection of hot water to about 75 °C for approximately 0.5-1 hours, then cooled to about 50-55 °C before entering the second reactor where enzymes were added (about 5 kg t⁻¹ MSW) resulting in hydrolysis and break-down of bonds in the organic materials, thereby suspending organic materials in the liquid phase (44). The retention time was about 10-16 hour. After the second

reactor, solid materials were separated from the liquid phase by a series of vibrating sieves. The liquid phase was further cleaned from small size "fluff" (e.g., pieces of cotton, textiles, plastic, and glass) by another vibrating sieve to produce the final bioliquid consisting of suspended organic materials (e.g., food waste, paper, and cardboard). The solid fraction separated from the liquid phase mainly consisted of non-degradable materials such as plastic, metals, textiles, soil, ceramics, etc.

The bioliquid was utilized for biogas production by anaerobic digestion. The solid fraction underwent further sorting for recovery of metal and plastic, while the remaining residual solids (including the "fluff") were incinerated for energy recovery. Overall, electricity and heat consumption were estimated to 25 kWh t⁻¹ MSW and 490 MJ t⁻¹ MSW, respectively.

Please refer to the SI for further details regarding the waste refinery process as well as all remaining technologies (i.e., incineration, conventional landfilling, bioreactor landfilling, mechanical-biological treatment, biological treatment, use-on-land, collection and transport, etc.).

Sensitivity and uncertainty analysis

Sensitivity and uncertainty analyses were addressed on two levels: i) scenario uncertainties and ii) parameter uncertainties. Uncertainties in the LCA methodology (e.g., characterization factors) were not addressed, as the implications of these were equal for all scenarios and best practices regarding LCA methodology were applied.

Scenario uncertainties were addressed by a number of sensitivity analyses: (S1) natural gas as marginal for electricity production (instead of coal as in the baseline); (S2) coal as marginal for heat production (instead of natural gas boilers as in the baseline); (S3) district heating produced at coal-fired CHP plants as marginal for heat (instead of natural gas boilers as in the baseline); (S4) no heat recovery (instead of heat recovery as in the baseline); (S5) diesel as marginal for transport (instead of gasoline as in the baseline); (S6) landfilling of the compost from bioliquid digestion (instead of use-on-land as in the baseline). S6 was included in order to emphasize the importance of N-leaching and soil contamination from use-on-land of compost derived from digestion of bioliquid produced at the waste refinery.

Parameter uncertainty and its implications on the ranking of scenarios was tested for two selected scenarios (*INC 0* and *WR GE 0*) by individually varying selected parameters: i) energy

content of the waste, ii) electricity efficiency of incineration, iii) electricity efficiency of biogas conversion, and iv) enzyme consumption in the waste refinery. Focus was to identify threshold values (i.e., break-even points) when the ranking of scenarios changed. The two scenarios *INC 0* and *WR GE 0* were selected as these scenarios represented the best performing scenarios with respect to GW (see discussion of results in section 3).

3. Results and discussion

The results of the LCA are presented in Figure 2 for the 42 baseline scenarios and the six sensitivity analyses (S1-S6 are only displayed where relevant: e.g., for the category Pres only S6 is relevant). The results are expressed as characterized impact potentials per tonne of wet MSW. Net contributions of each process (e.g., recycling or incineration) and total impacts for each scenario were obtained by subtracting the avoided impacts (negative values in the figures) from the induced impacts (positive values). Net values below zero indicate overall environmental benefits. The discussion is focused on management of the residual waste (rMSW) and the organic waste as the contributions from source-segregation of aluminum, ferrous metals, plastic, paper, and glass were identical for all scenarios with a specific waste composition (i.e., *a* and *b* scenarios). Details regarding energy and waste materials balances can be found in the SI.

Environmental performance without organic source-segregation (scenarios 0)

For GW and AC, the waste refinery performed similar to incineration and MBT (involving anaerobic digestion and CHP) when the biogas was used for CHP. Among these scenarios, the relative difference in greenhouse gas (GHG) savings was small (about 2-14%). The final GW ranking was determined by the waste composition (*a* and *b*): For low-organic waste, incineration (*INC*) performed best while for high-organic waste, the waste refinery (*WR GE*) achieved larger savings. Scenarios with lower energy recovery (*CLF* and *MBT DC*) always performed worse with respect to GW and AC, reflecting that the overall energy conversion efficiencies of incineration and the waste refinery were better (Figure S16). The waste refinery had the highest electricity recovery for both waste types (*a* and *b*) while incineration by far provided the largest district heating production (Figure S16).

The waste refinery scenarios involving upgrading of the biogas to transport fuel (*WR TF*) generally showed lower GW savings primarily as a consequence of the lower CO₂ emissions

associated with the substituted fuel (gasoline) compared with coal in case of electricity. However, when natural gas was the marginal fuel for electricity, utilizing biogas for CHP and transport was comparable (see section ” Scenario and parameter uncertainties”).

Phosphorous recovery was maximized in the waste refinery scenarios (*WR GE* and *WR TF*) due to the potential for nutrient utilization on farmland, estimated to 85% of the P content in the input waste. This, however, came at the expense of eutrophication (EP (N)) caused by potential N leaching (estimated to 20% of the N applied) and of toxic impacts (ETw and HT) caused by transfer of heavy metals to the bioliquid and subsequently to the compost .

Effects of organic source-segregation (scenarios *I* and *II*)

Organic source-segregation was not needed for the waste refinery, as the key focus here was to transfer organic materials into the bioliquid. Source-segregation therefore did not improve the environmental performance of the waste refinery. However for GW, source-segregation improved the overall electricity recovery in the incineration (*INC*) and MBT scenarios (*MBT AC*) in the case of high-organic waste (*b*) as anaerobic digestion provided higher electricity recovery (see section ”Effects of the waste composition”). Both technologies, however, were comparable with the waste refinery (*WR GE*). Scenarios with no or partial energy recovery from rMSW (*CLF* and *MBT DC*) significantly increased their overall GW and AC savings due to the energy recovery introduced from biological conversion of the segregated organic waste. This was expected, and confirmed previous studies (39). No significant effects from organic source-segregation were observed for EP (N) as NH_3 and NO_3^- emissions were comparable for the two levels of source-segregation.

Higher organic source-segregation efficiencies (exemplified by 100% in scenario *II*) would primarily benefit scenarios with no or only partial energy recovery from rMSW (*CLF* and *MBT DC*), incineration (*INC*) and MBT with combined treatment (*MBT AC*) in case of high-organic waste composition (*b*). Although P savings were largest in the waste refinery scenarios, P savings were increased in all remaining scenarios (up to about 56% of P content of the input waste) when organic source-segregation was introduced.

Effects of the waste composition

Comparing scenarios *a* and *b*, it can be seen that the waste composition significantly affected the ranking of technologies and the magnitude of savings, mainly for GW, ETw, HT, Pres, and scenarios with no source-segregation (*0*).

Energy recovery from the waste refinery was determined by the content of organic materials, paper, and cardboard in the waste and therefore significantly affected by changes in waste composition, including water content of the materials. Similarly for MBT and incineration. This was emphasized by the energy balance shown in Figure S16: without organic source-segregation (*0*), the electricity recovery from the waste refinery (*WR GE*) was always higher than incineration. The difference between electricity produced in the *WR GE* and *INC* scenarios was significantly larger in *b* (40%) compared with *a* (15%); the reason was the larger amount of rMSW and fraction of organic waste in rMSW in *b*, see Table S1.

Heat production in the waste refinery scenarios was lower than incineration (-19% in *b* and -30% in *a*). Combined with the environmental impacts from the refining process itself (energy and enzyme consumption), the waste refinery (*WR GE*) performed worse than incineration (*INC*) in GW and AC with waste *a*. This was not the case for waste *b* where the higher share of organic waste provided more biogas and higher electricity production, largely compensating for the refining process.

For ETw and HT, the differences in waste composition affected the magnitude of the impacts associated with use-on-land of compost. The impacts were considerably larger for low-organic waste (*a*) as a result of the higher concentration of heavy metals in the waste (30) compared with (23). For the high-organic waste (*b*), more bioliquid was obtained from organic waste in the waste refinery, resulting in lower (i.e., more diluted) concentrations of metals in the bioliquid (see Table S1 and S5).

The toxic impacts (ETw and HT) did not decrease in the waste refinery scenarios as an effect of organic source-segregation (*I* and *II*) as toxic metals (Hg, Ni, Cd, Cu, etc.) were not transferred to the bioliquid from organic waste materials but rather from materials such as paper, cardboard, cat litter, cartons, metals, dust, vacuum cleaning bags, etc. These fractions, even with organic source-segregation, would still be included in the rMSW and sent to the waste refinery. Higher fractions of organic materials in waste *b* resulted in larger P resource savings compared with waste *a*. Overall, the best environmental performance of the waste refinery was observed for the high-organic waste.

Use-on-land of waste refinery residues

Toxic impacts associated with use-on-land of the compost after digestion of bioliquid from the waste refinery can be considered a main concern. Potential options for minimizing metal release to soil may be: i) post-treating the bioliquid-digestate (before composting) to allow selective P recovery by precipitation, or simply avoiding use-on-land by ii) landfilling the compost or iii) incinerating the digestate after dewatering. The latter two options result in less nutrients being recovered. While LCA modeling focuses on potential toxic impacts by aggregating loads of contaminants being emitted to the environment, risk assessment should be applied to address specific effects on soil, plants, animals, and humans related to actual concentration levels. As such, the LCA results indicate that site-specific assessment of use-on-land of the products should be carried out.

Scenario and parameter uncertainties

The sensitivity analyses highlighted that assumptions regarding the energy system were crucial, both for electricity and district heating. When the marginal fuel for electricity was natural gas (S1), the GHG savings associated with energy recovery were considerably decreased, affecting the performance of all the scenarios and finally changing the ranking between them. Both the performance of the bioreactor landfill (*BLF*) and the waste refinery transport fuel scenario (*WR TF*) were significantly improved (relative to the other scenarios). In this situation, C sequestration became a main contributor to GHG savings, thereby ranking the bioreactor landfill highest among the others. This was in accordance with other studies identifying C sequestration as important (39, 45-48). With natural gas as the marginal for electricity, utilization of biogas from the waste refinery (either CHP or transport fuel) became comparable with respect to GW. As provision of transport biofuels in the future may be challenging and ultimately induce more energy crop production (4), waste-derived biofuels may offer a valuable alternative without upstream impacts from land use changes (3, 5, 6).

With coal as marginal for heat production (S2), incineration always performed best. This was attributed to increased benefits (GHG, SO₂, and NO_x) associated with the avoided emissions from coal-based heat production. When heat was assumed not recovered in all scenarios (S4), bioreactor landfilling performed best relative to GW while the ranking of the remaining

scenarios did not change. GW savings from incineration (*INC*), MBT with combined treatment (*MBT AC*), and waste refinery with CHP (*WR GE*) were comparable as was the case for the baseline.

Substitution of diesel instead of gasoline (*S5*) did not significantly affect the performance of the transport scenario (*WR TF*). Landfilling of compost produced from the bioliquid (*S6*) strongly decreased the potential N-eutrophication and toxic impacts. On the other hand, P recovery was essentially cancelled.

The parameter uncertainty analysis emphasized the crucial importance of the waste composition for the ranking between incineration (*INC 0*) and waste refining (*WR GE 0*) relative to GW (Figure S17). For the low-organic waste (*a*), only low electricity efficiency of the incinerator (<15%) and/or high electricity efficiency of the biogas-to-electricity conversion (>50%) could make the waste refinery environmentally favorable. However, for high-organic waste (*b*), the electricity efficiency of incineration should be higher than 32% to balance the GW savings from the waste refinery.

Implications for future selection of waste technologies

Overall, the waste refinery provided comparable GW savings as state-of-the-art incineration, MBT, and bioreactor landfilling. On the other hand, the waste refinery allowed significantly higher recovery of P resources, although at the likely consequence of additional toxic emissions and loss of N by use-on-land. While waste refining may still evolve as a concept and the technology be further optimized in the future, the results clearly indicate that no waste technology can be singled out as environmentally preferable. Provided that technology implementation is state-of-the-art.

The results suggest that decision-makers should avoid having a too narrow focus on GW performance, but also address other aspects such as economical performance, social acceptance, local involvement, technical robustness, etc. Decision-makers should consider to which extent phosphorous resources in waste should be prioritized, e.g., at the expense of toxic emissions. And regarding energy, whether waste-derived transport fuels should be preferred over electricity generation.

While the waste refinery cannot be considered a clear winner overall, two aspects are crucial for future selection: i) the waste composition and ii) society's needs for the outputs from

waste. The boundary conditions for the waste management system, e.g., local traditions, society structures, consumer behavior, etc., thereby become decisive.

Supporting Information (SI)

Additional information on: system boundaries, marginal energy technologies and fertilizers, LCA process flow diagrams, waste composition, LCI of waste technologies, waste material and energy balances, and sensitivity analyses is available free of charge via the Internet at: <http://pubs.acs.org>.

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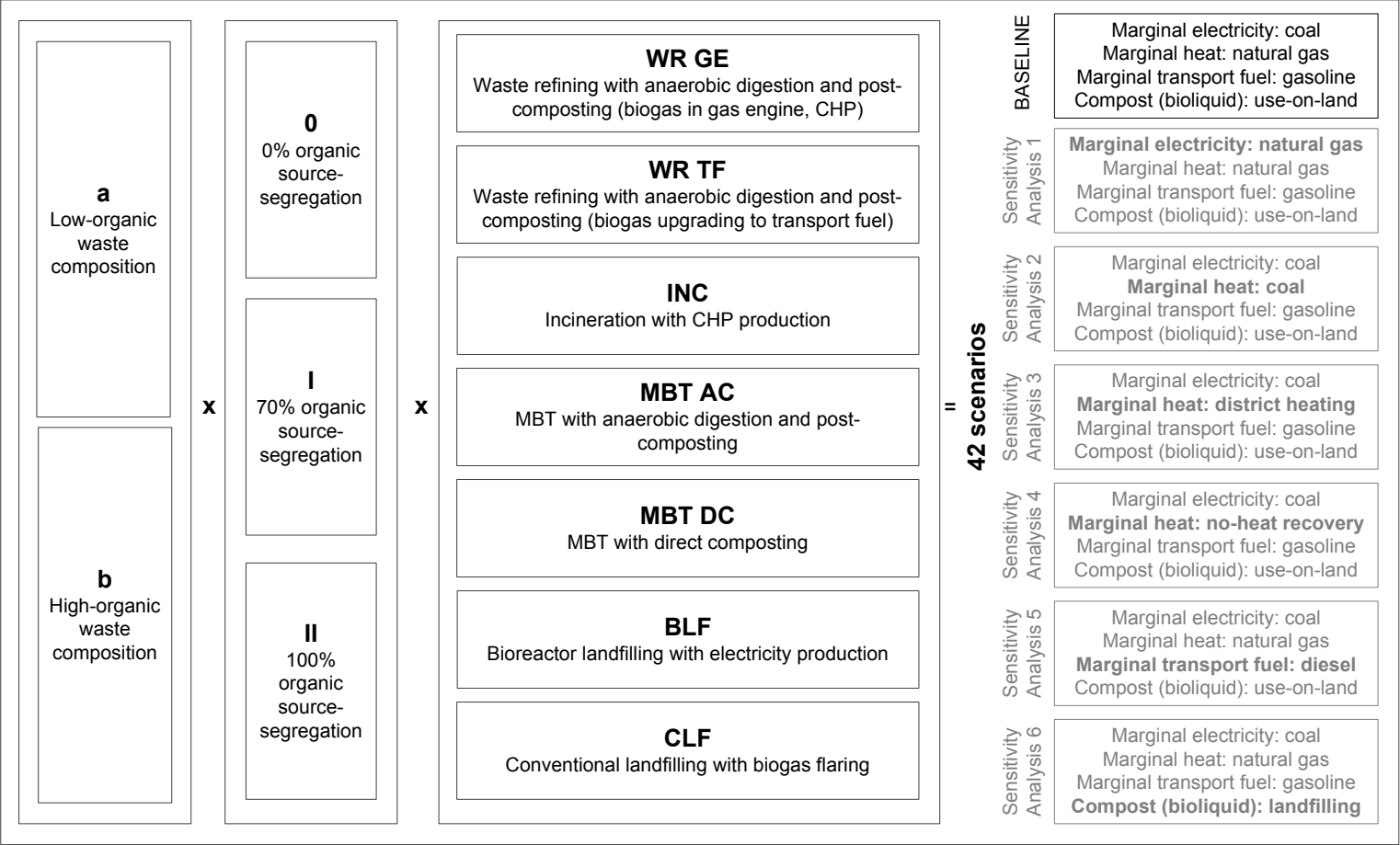
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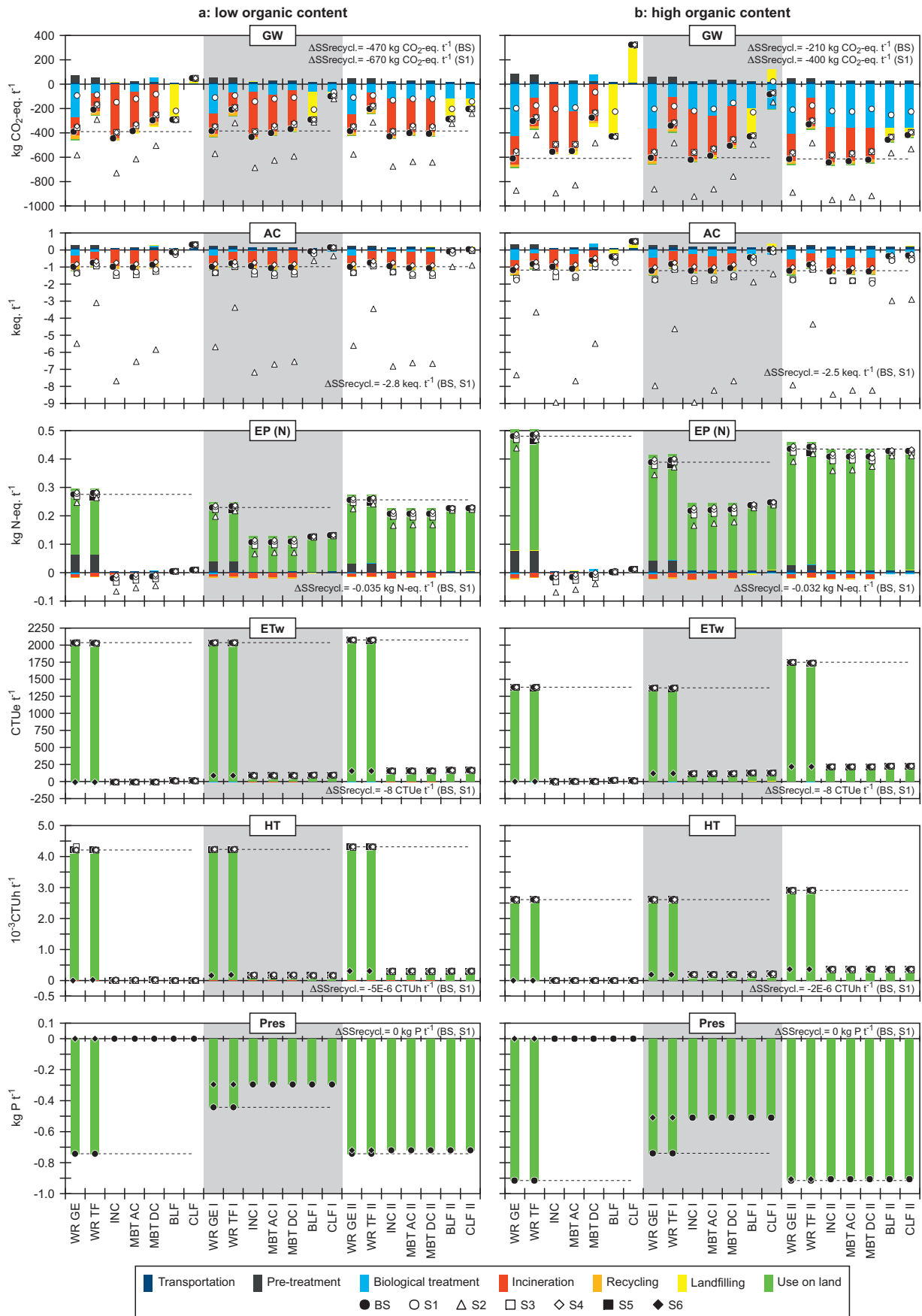
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Supporting Information (SI)

Environmental Science & Technology

Material Resources, Energy, and Nutrient Recovery from Waste: Are Waste Refineries the Solution for the Future?

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This SI document includes text, tables, and figures with details on the process data for the inventory analysis of the LCA. Additional information on waste materials and energy balances, and sensitivity analyses are also provided.

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1. LCA scenarios modelling and system boundary

This section provides additional information with respect to the modelling and system boundary of the investigated scenarios. As thoroughly detailed in the manuscript a total of 252 scenarios have been addressed. Figure S1-S14 illustrates the boundary conditions for the scenarios 0 and I for the case of waste composition (a) (only relevant flows; values are rounded to two significant digits). Notice that electricity and heat produced are net values (i.e., plants own consumptions have been subtracted, including energy for pre-treatments). The modelling for the case of scenarios II and for the case of waste composition (b) is similar. For the latter, additional information on waste materials and energy balances can be found in section 4. Table S3 provides an overview of technologies and pre-treatments involved in the scenarios under assessment.

1.1 Waste composition

As detailed in the main manuscript, two sets of waste compositions (*a* and *b*) were used in the assessment. These are shown in Table S1-S2 along with the composition of the residual MSW (rMSW representing the left-over after source-segregation of recyclable materials and organic waste) in the assessed scenarios 0, I, and II. Details regarding waste composition, water content, and chemical composition of the individual waste material fractions can be found in (1, 2). For details regarding amounts of individual material fractions, source-segregated MSW (ssMSW), and residual MSW (rMSW), please refer to section 4 and Table S9. Please note that not all individual waste materials within waste type mentioned in Table S2 were assumed source-segregated, e.g., dirty paper and dirty cardboard were considered routed to the rMSW for further treatment. The share of dirty paper was estimated to ca. 1.9% (of the total) in waste (a) and to 1.4% (of the total) in waste (b). Dirty cardboard was 0.98% in (a) and 1.4% in (b). For plastic, non-recyclable plastic items (e.g., plastic toys) were estimated to 5.1% in (a) and 1.5% in (b). The low share of recyclable plastic in (a) can be largely attributed due to the Danish return system (in Denmark a large share of the plastic bottles are collected through a separate collection-refunding system, and were not considered as part of the generated household waste in the investigations performed by (1), i.e. not part of the household-bin). For glass, the non-recyclable share was 0.24% in (a) and 0.82% in (b). All materials in the fractions ferrous metal and aluminium (Table S1-S2) were considered recyclable. As an example, the amount of paper source-segregated in the case of waste (a) would equal the amount of clean paper multiplied by the separation efficiency (0.8), i.e., on

1000 kg MSW: $(250 \text{ kg} - 19 \text{ kg}) \cdot 0.8 = 184.8 \text{ kg}$ (180 kg with two significant digits, as reported in Table S9 and Figures S1-S14).

Table S1. Overview of the MSW composition datasets used in the study; (a): (1); (b): (2). The rMSW composition (as modelled after source-segregation of selected recyclables and organic waste) is also presented; rMSW 0: no organic source-segregation; rMSW I: organic source-segregation (efficiency 70%); rMSW II: organic source-segregation (efficiency 100%). Any inconsistencies are due to rounding (values are rounded to two significant digits).

Waste material fraction	MSW		rMSW 0		rMSW I		rMSW II	
	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)
1. Organic waste	37	60	54	72	30	45	9.5	8.4
2. Paper	25	9.6	9.4	3.7	15	7.1	19	12
Clean paper	23	8.2						
Dirty paper	1.9	1.4						
3. Cardboard	5.7	4.1	2.8	2.3	4.3	4.5	5.6	7.6
Clean cardboard	4.7	2.7						
Dirty cardboard	0.98	1.4						
4. Cartons and alike	2.4	1.2	3.4	1.4	5.3	2.8	6.8	4.7
5. Plastic	6.9	9.3	8.8	6.5	13	13	17	21
Recyclable plastic	1.8	7.8						
Non-recyclable plastic	5.1	1.5						
6. Textile	1.8	3.5	2.6	4.2	4.0	8.2	5.1	14
7. Glass	9.0	3.3	3.3	1.0	5.0	1.9	6.5	3.2
Recyclable glass	8.8	2.5						
Other glass	0.24	0.82						
8. Ferrous metal	1.6	2.5	0.57	0.75	0.88	1.5	1.1	2.4
9. Aluminium	0.76	0.50	0.28	0.15	0.42	0.29	0.55	0.49
10. Other	10	6.6	14	8.0	22	15	29	26
Total (1 to 10)	100	100	100	100	100	100	100	100

Table S2. Overview of the waste material fractions considered in the assessment. Lower heating values (LHV) assumed for waste materials as generated are provided as MJ kg DM⁻¹ for waste (a) and (b).

Macro-waste material	LHV (a/b)	Waste material fractions
Organic waste	19.1/15	Vegetable food, animal food, kitchen tissues, wood-like materials (yard waste)
Paper	15.4/14.7	Newsprints, magazines, advertisements, books and phonebooks, office paper, other clean paper, dirty paper
Cardboard	17.2/14.7	Cardboard containers, dirty cardboard
Cartons and alike	22/23.6	Milk cartons and alike, cartons with aluminium foil
Plastic	33.3/38.5	Soft plastic, plastic bottles, hard plastic, non-recyclable plastic
Textile	20.7/23	Textiles, shoes, and leather
Glass	0.0/0.0	Clear glass, green glass, brown glass, other glass
Ferrous metal	0.0/0.0	Metal containers, other mixed ferrous metals
Aluminium	3.7/3.7	Aluminium containers and tins, aluminium trays/foils
Other	15/28.9	Office articles, cigarettes butts, diapers, sanitary towel, bandages, cotton, tampons, vacuum cleaner bags, soil, stones, batteries, animals gravel and litter, ceramics, ashes, plastic-coated aluminium foils, other non-combustibles

Figures S1-S14 display the main (selected) mass and energy flows involved in the scenarios under assessment. Please, notice that the amount of aluminium (AL) recovered from bottom ash accounts for all the aluminium (Al) that is found in the waste incinerated and not only for the Al found in the non-segregated share of the macro-fraction “Aluminium” reported in Table S1-S2 (for example plastic-coated aluminium foil and cartons with aluminium foil also contain Al). In addition, recovery of ferrous metal and aluminium from bottom ash was considered for all scenarios involving incineration (i.e., WR GE, WR TF, INC, MBT AC, MBT DC), although this is not specifically displayed for WR GE, WR TF, MBT AC, and MBT DC due to space limitation in the associated Figures. In these, the recovery of ferrous metal and aluminium from incineration bottom ash corresponded to ca. 0.4 kg and 0.2 kg, respectively.

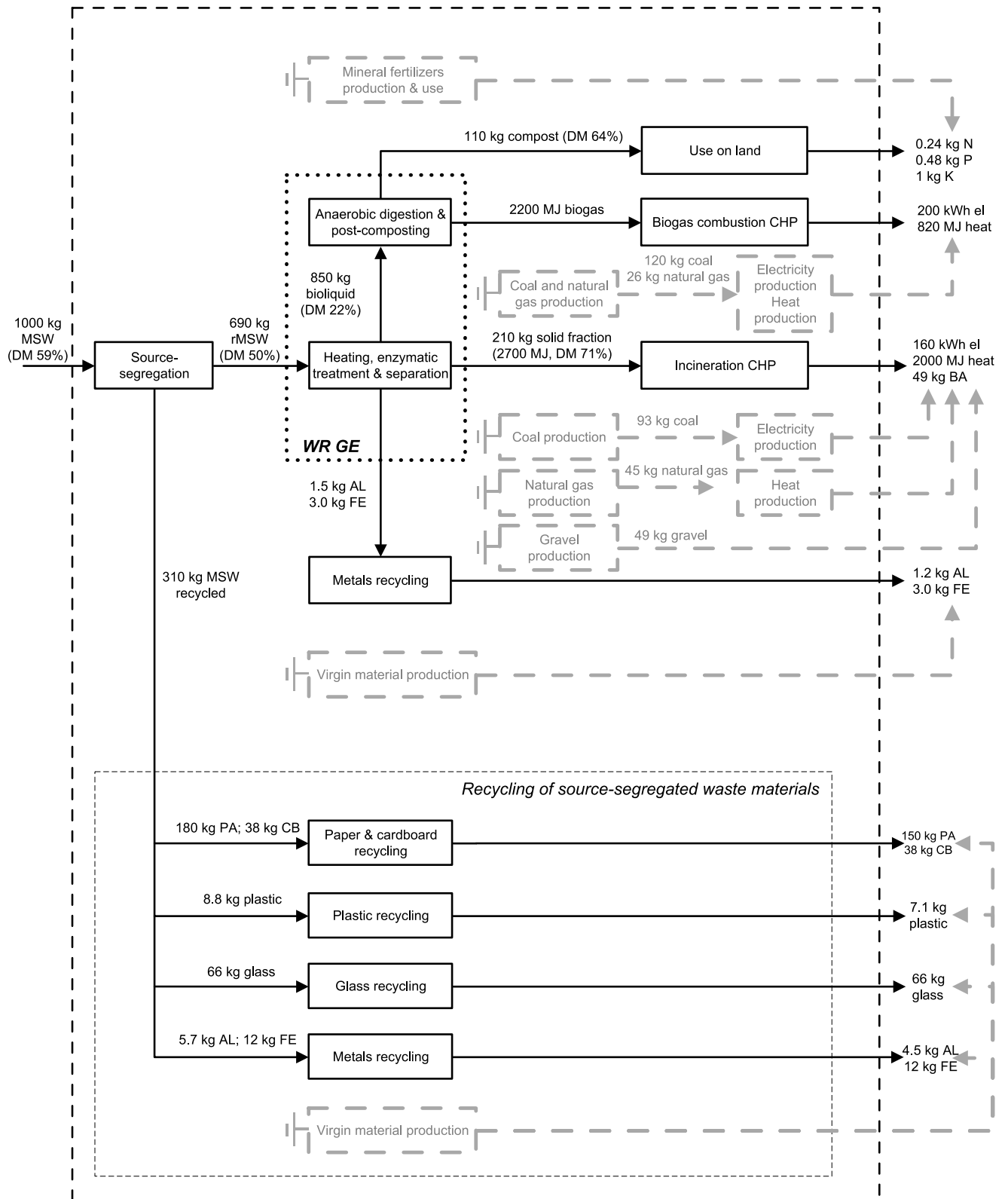


Figure S1. LCA system boundary of WR GE 0 for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

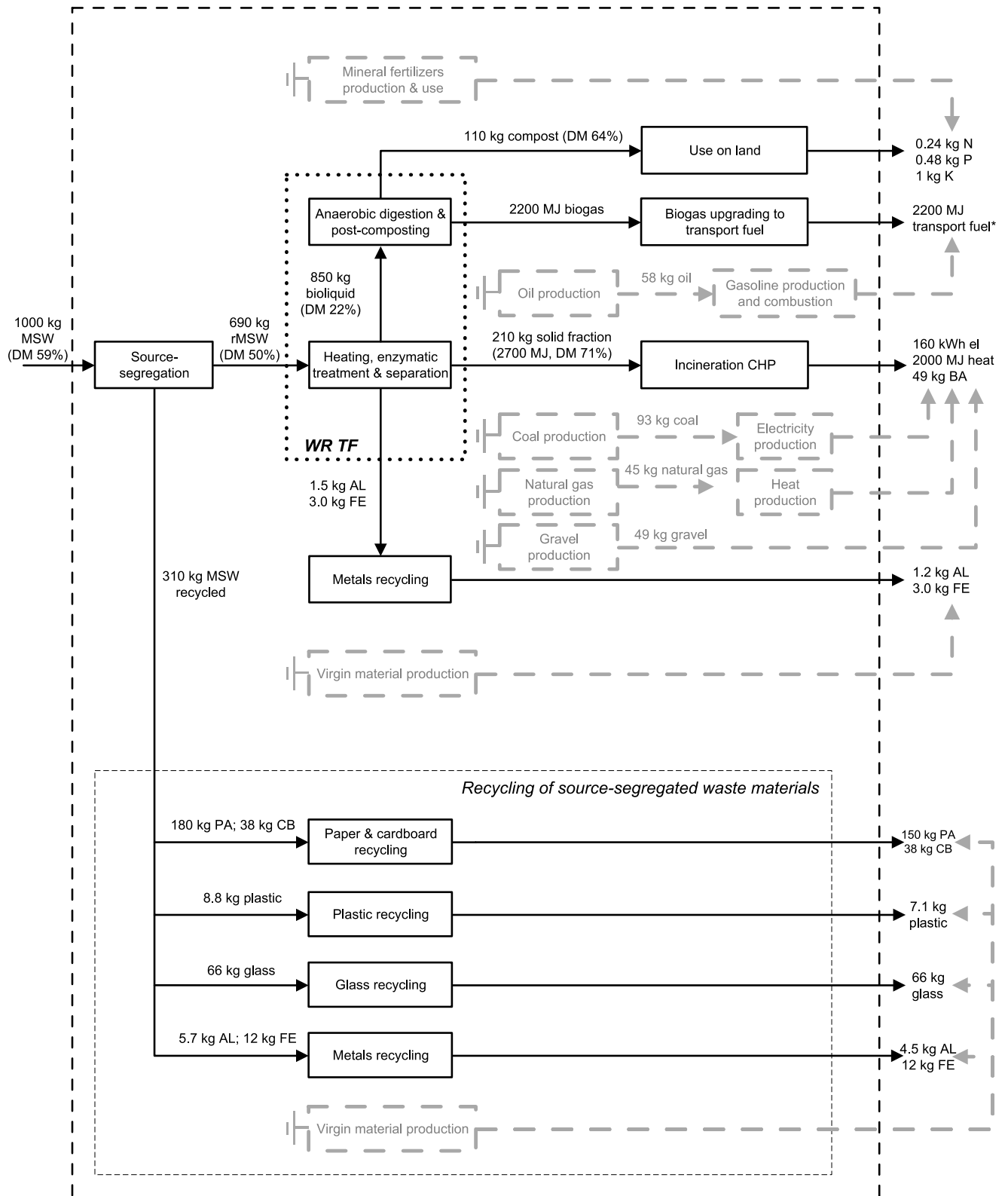


Figure S2. LCA system boundary of WR TF 0 for the case of waste composition (a). *The energy consumed for the upgrading process corresponded to $0.014 \text{ kWh MJ}^{-1} \text{ CH}_4$ (not visualized). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

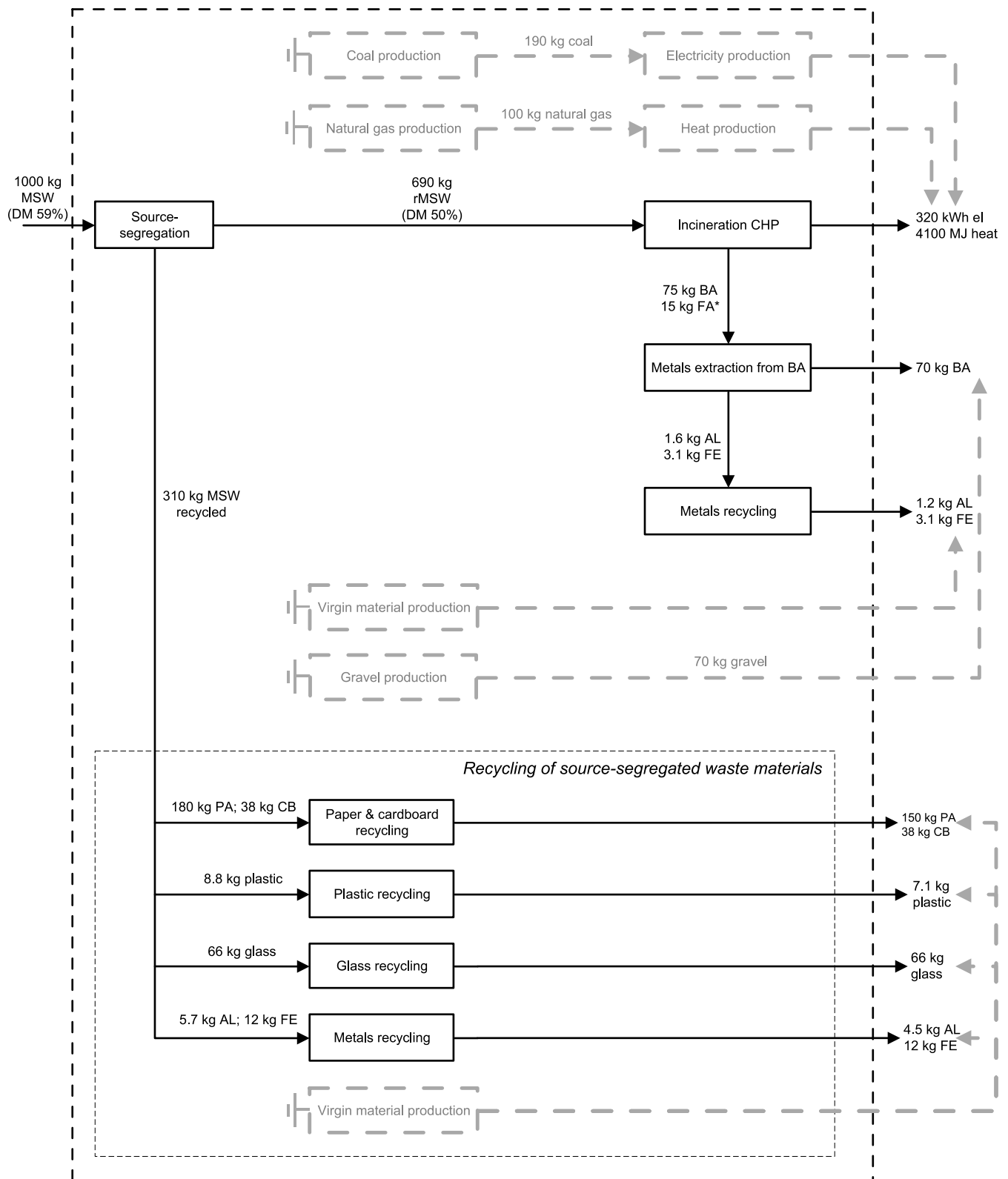


Figure S3. LCA system boundary of INC 0 for the case of waste composition (a). AL: aluminium; BA: bottom ash; CB: cardboard; CHP: combined-heat-and-power; FA: fly ash; FE: ferrous metal; PA: paper; rMSW: residual MSW. *Used for backfilling of old salt mines.

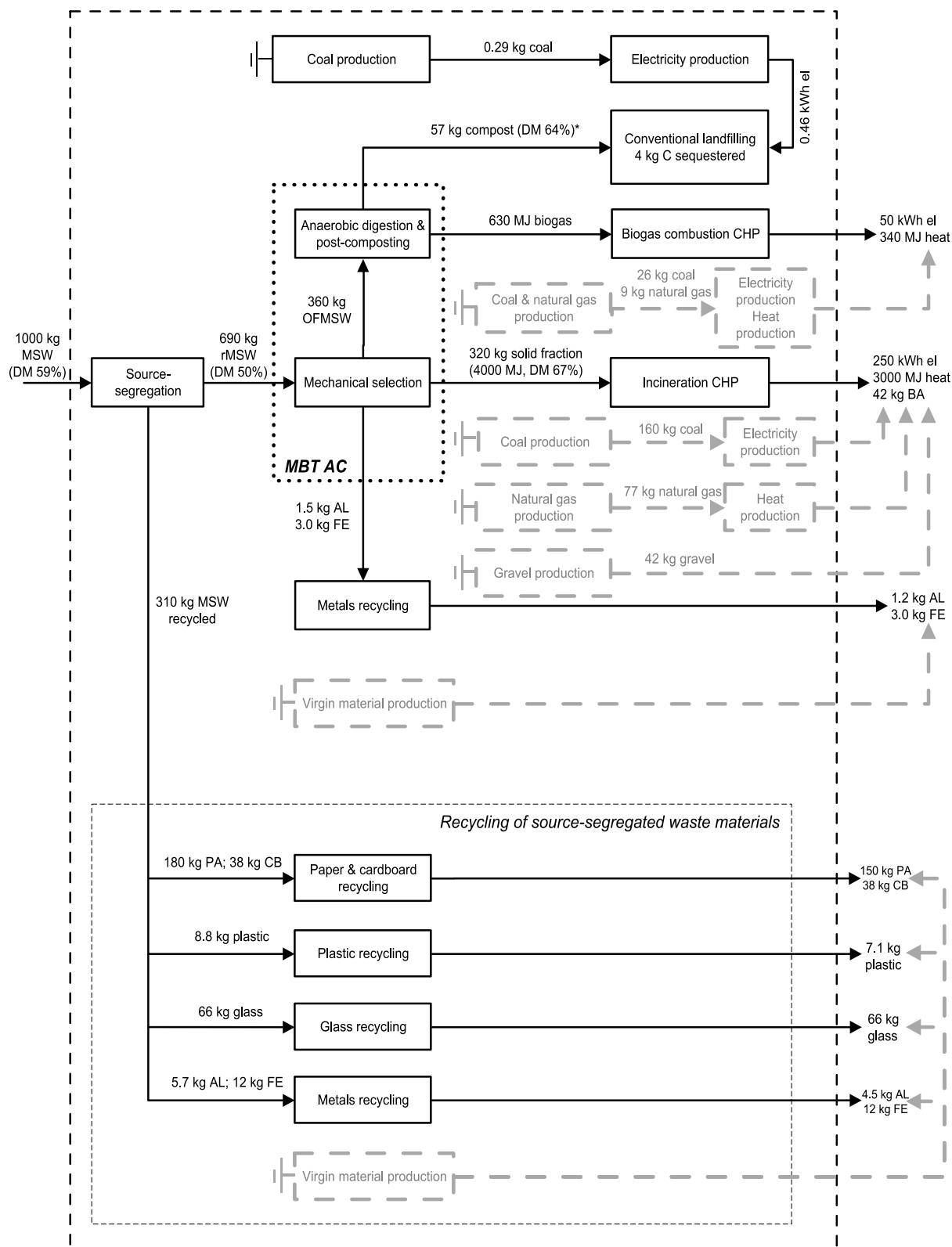


Figure S4. LCA system boundary of MBT AC 0 for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

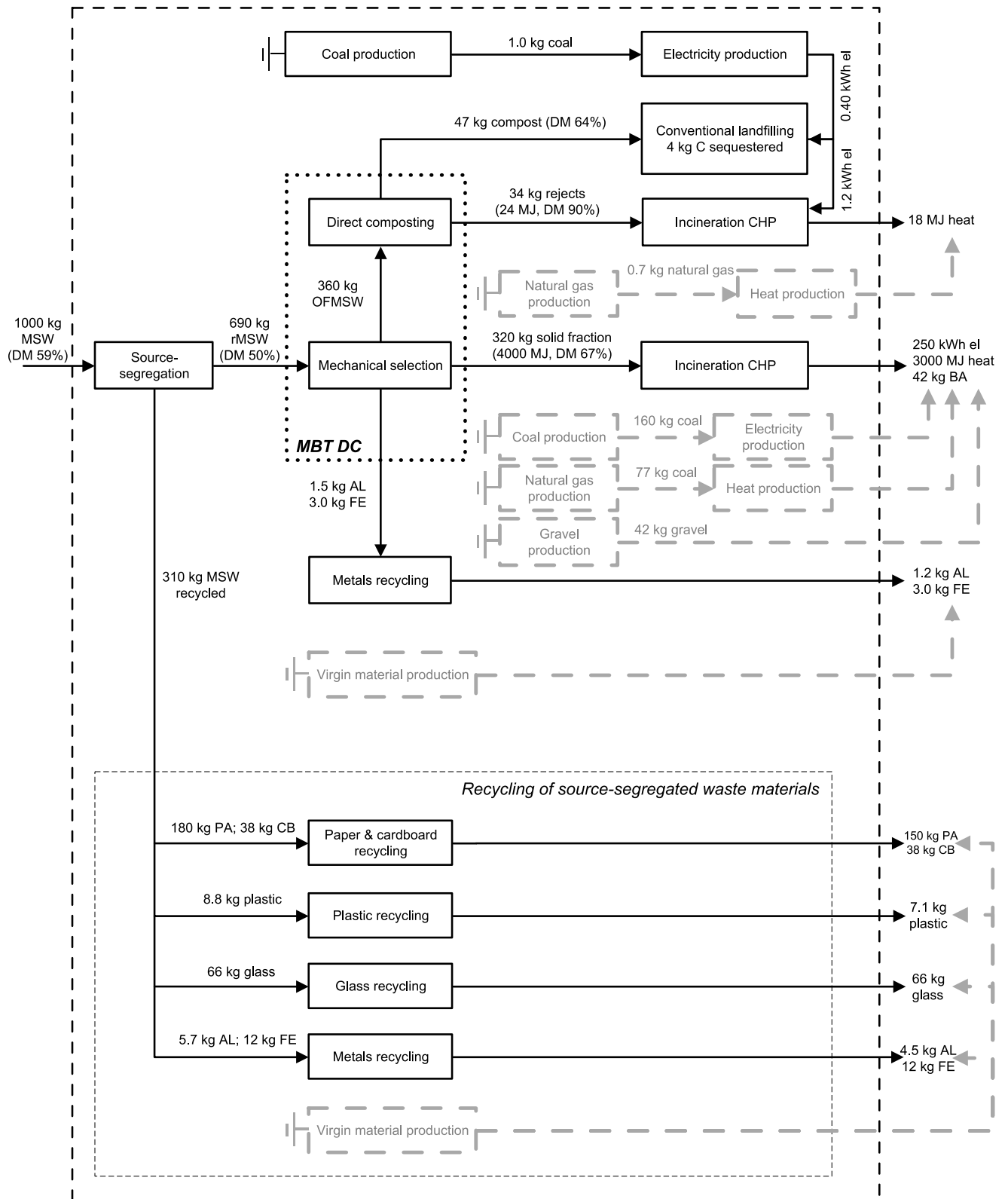


Figure S5. LCA system boundary of MBT DC 0 for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

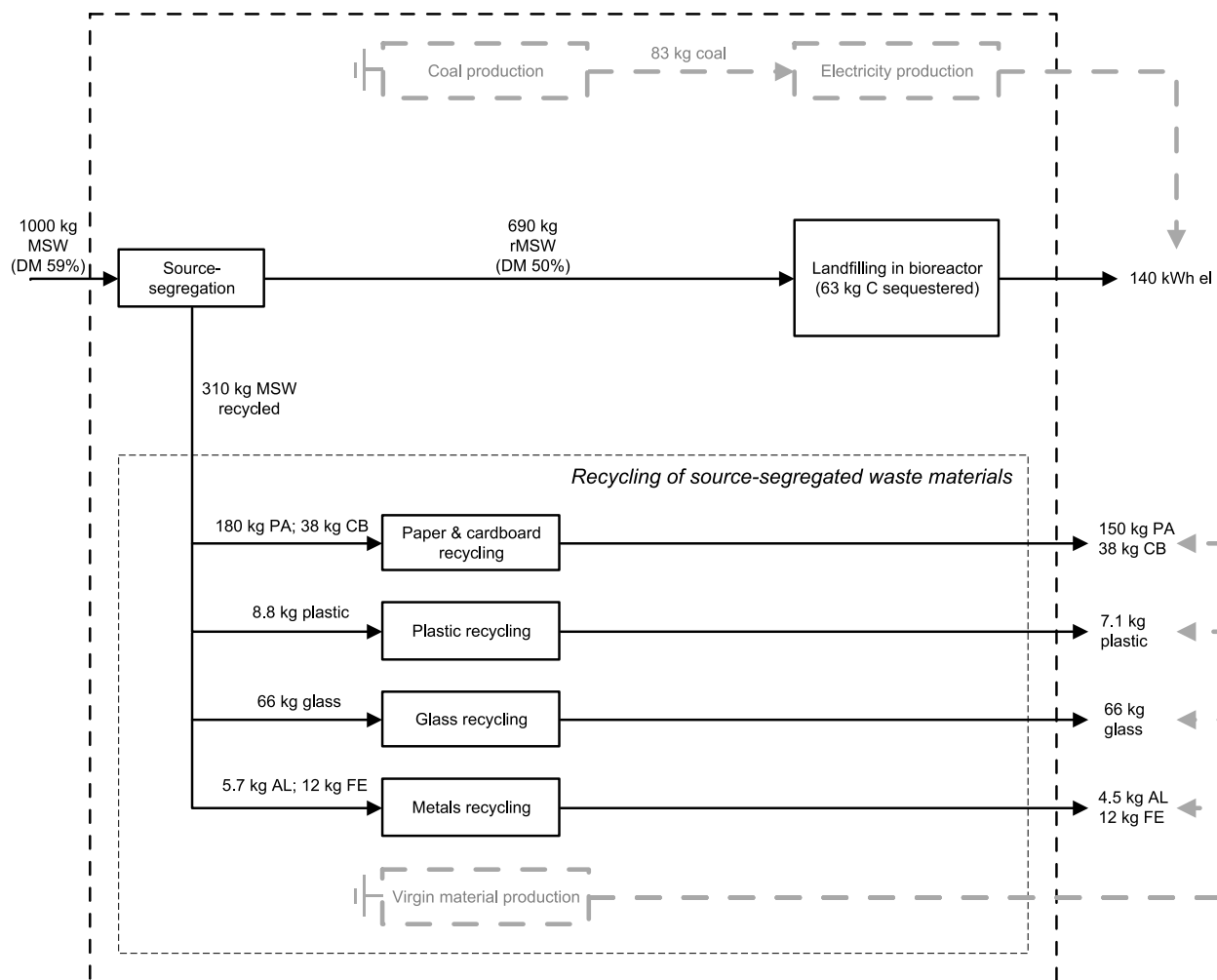


Figure S6. LCA system boundary of BLF 0 for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

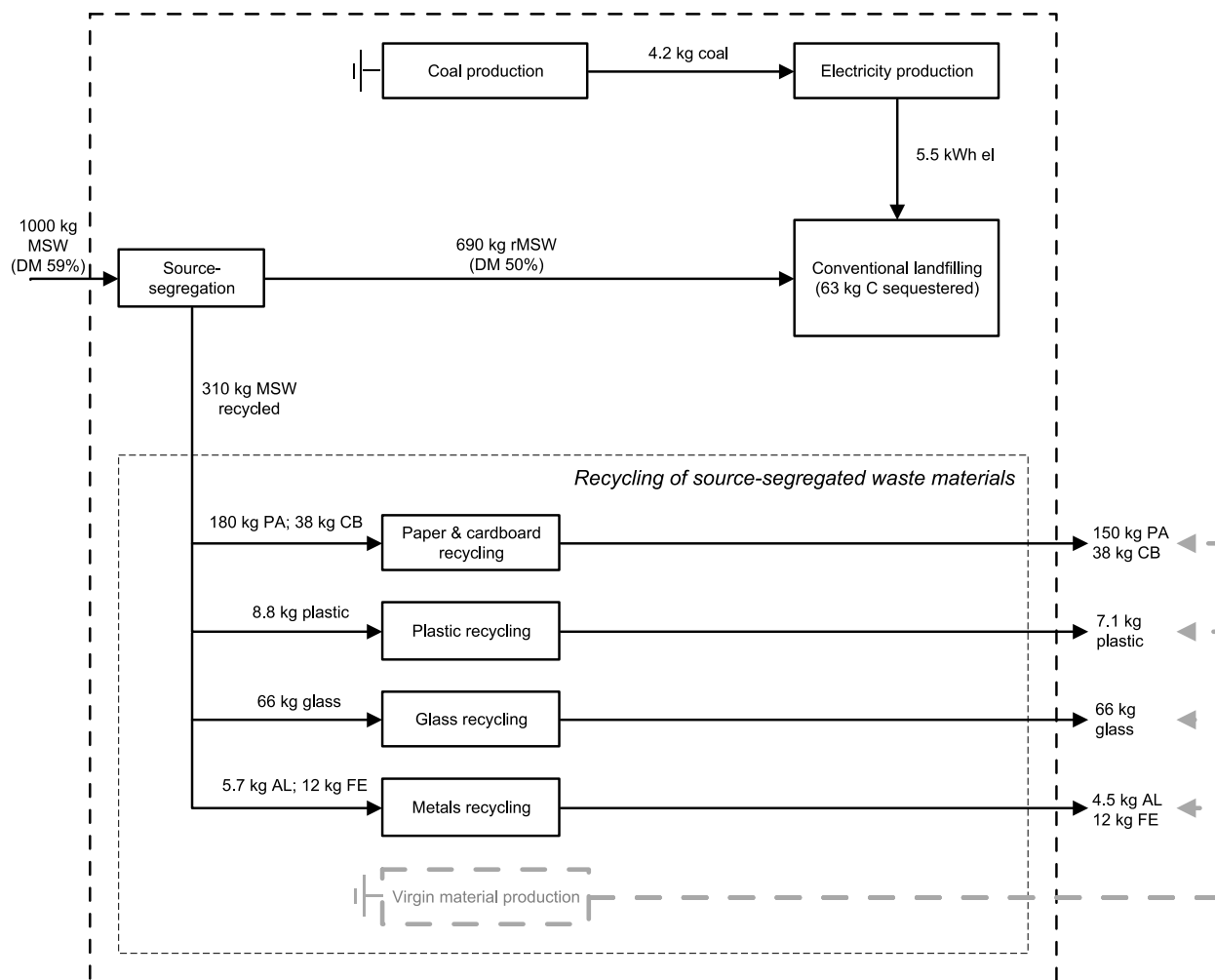


Figure S7. LCA system boundary of CLF 0 for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

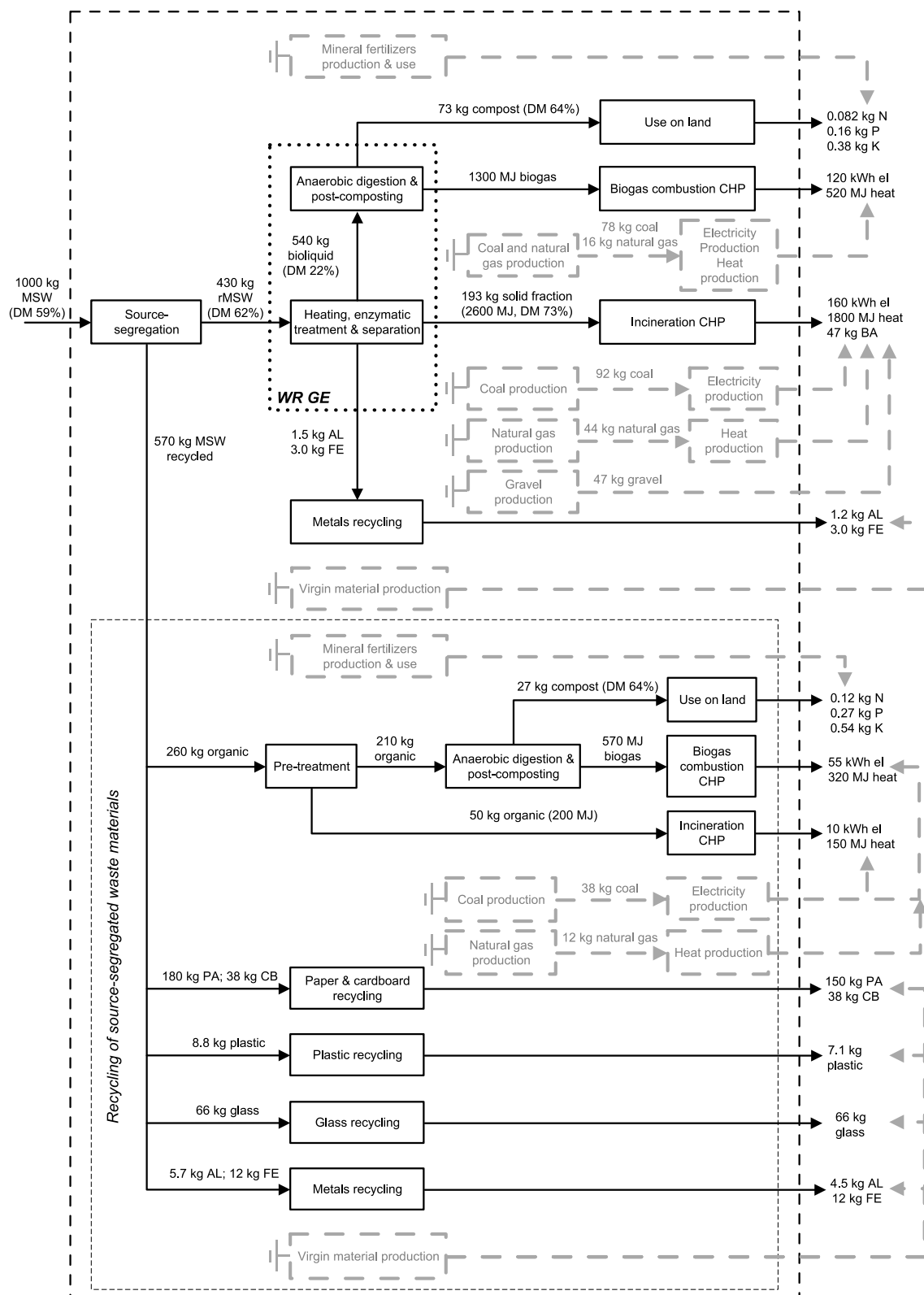


Figure S8. LCA system boundary of WR GE I for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

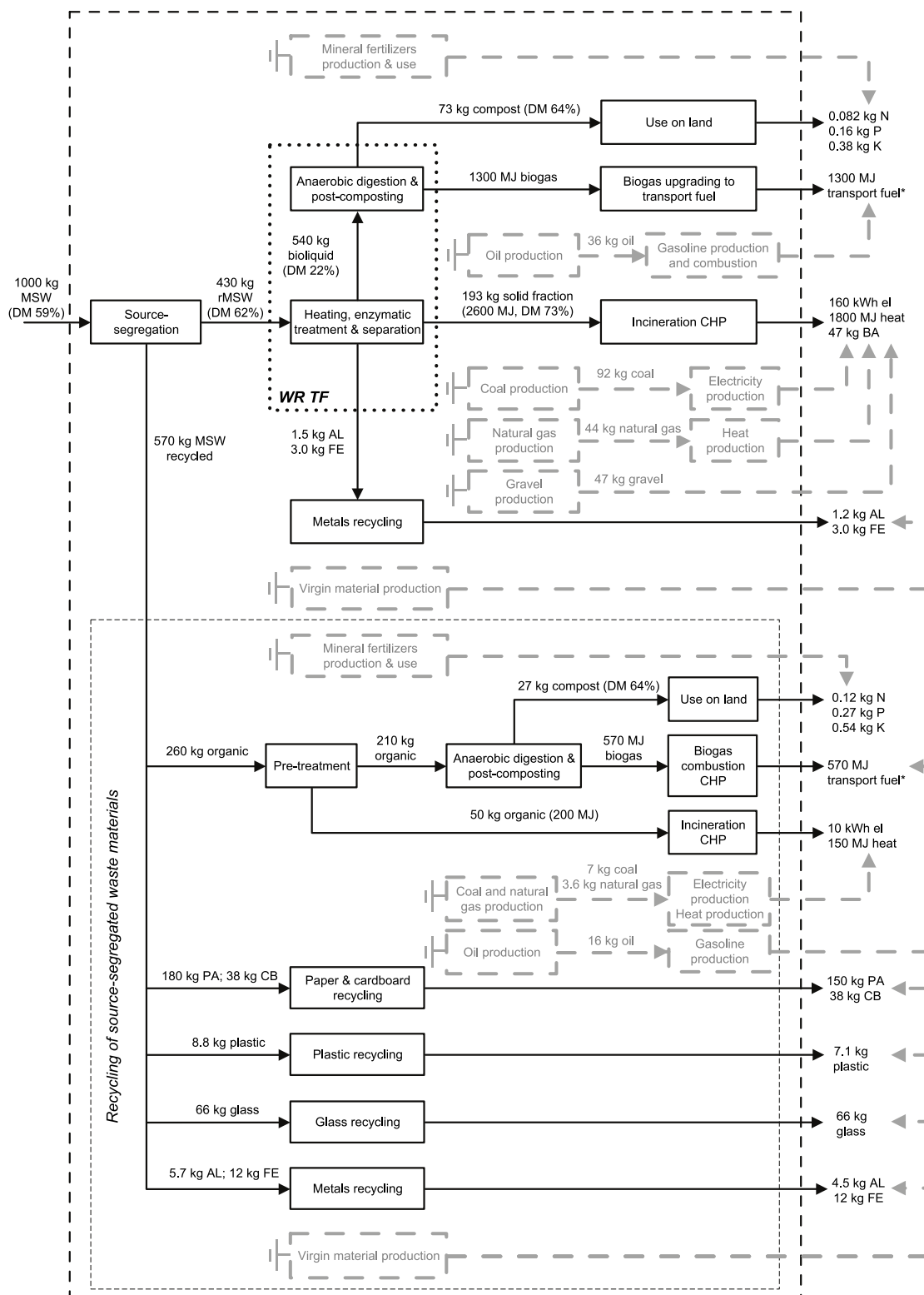


Figure S9. LCA system boundary of WR TF I for the case of waste composition (a). *The energy consumed for the upgrading process corresponded to $0.014 \text{ kWh MJ}^{-1} \text{ CH}_4$ (not visualized). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

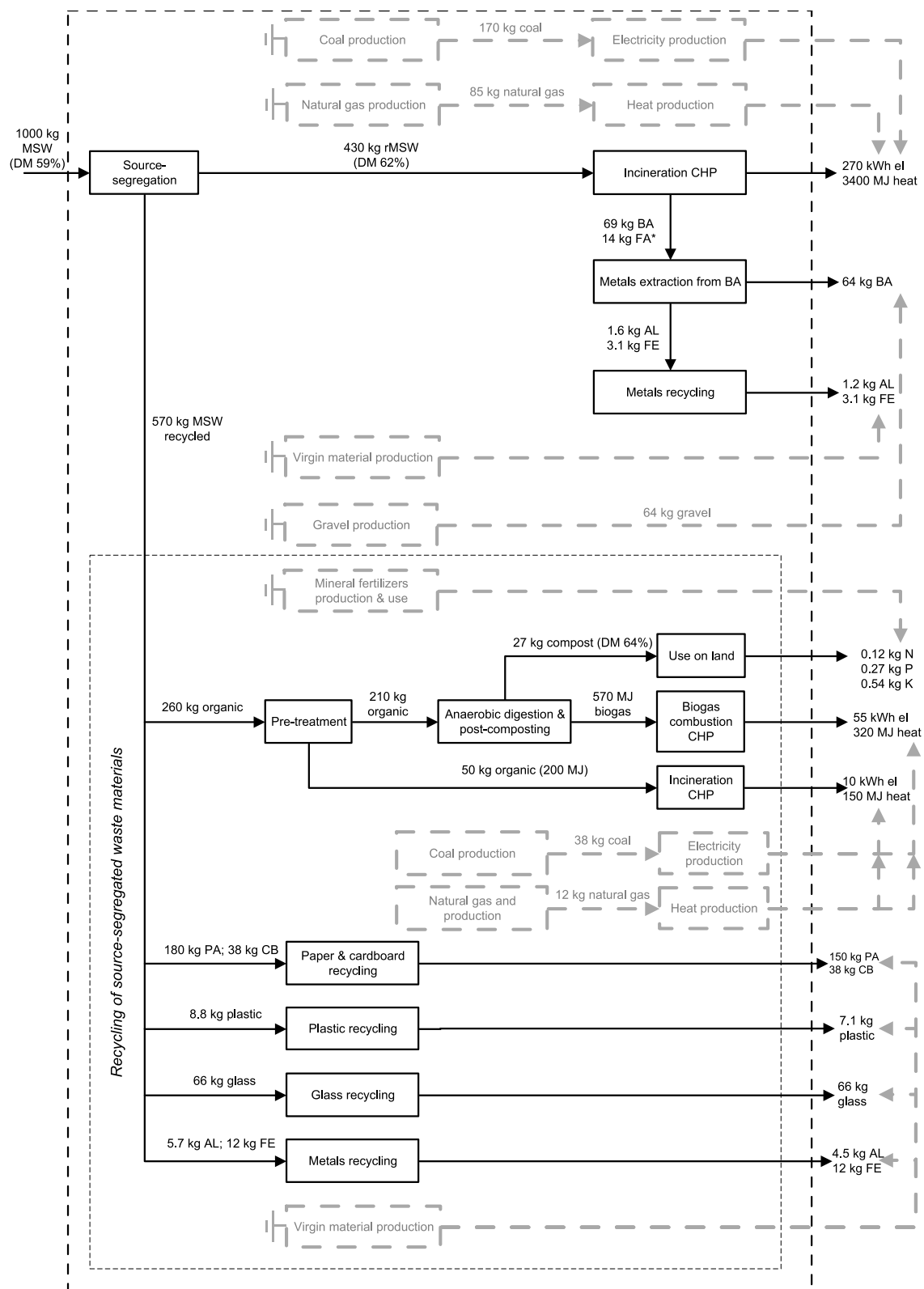


Figure S10. LCA system boundary of INC I for the case of waste composition (a). AL: aluminium; BA: bottom ash; CB: cardboard; CHP: combined-heat-and-power; FA: fly ash; FE: ferrous metal; PA: paper; rMSW: residual MSW. *Used for backfilling of old salt mines.

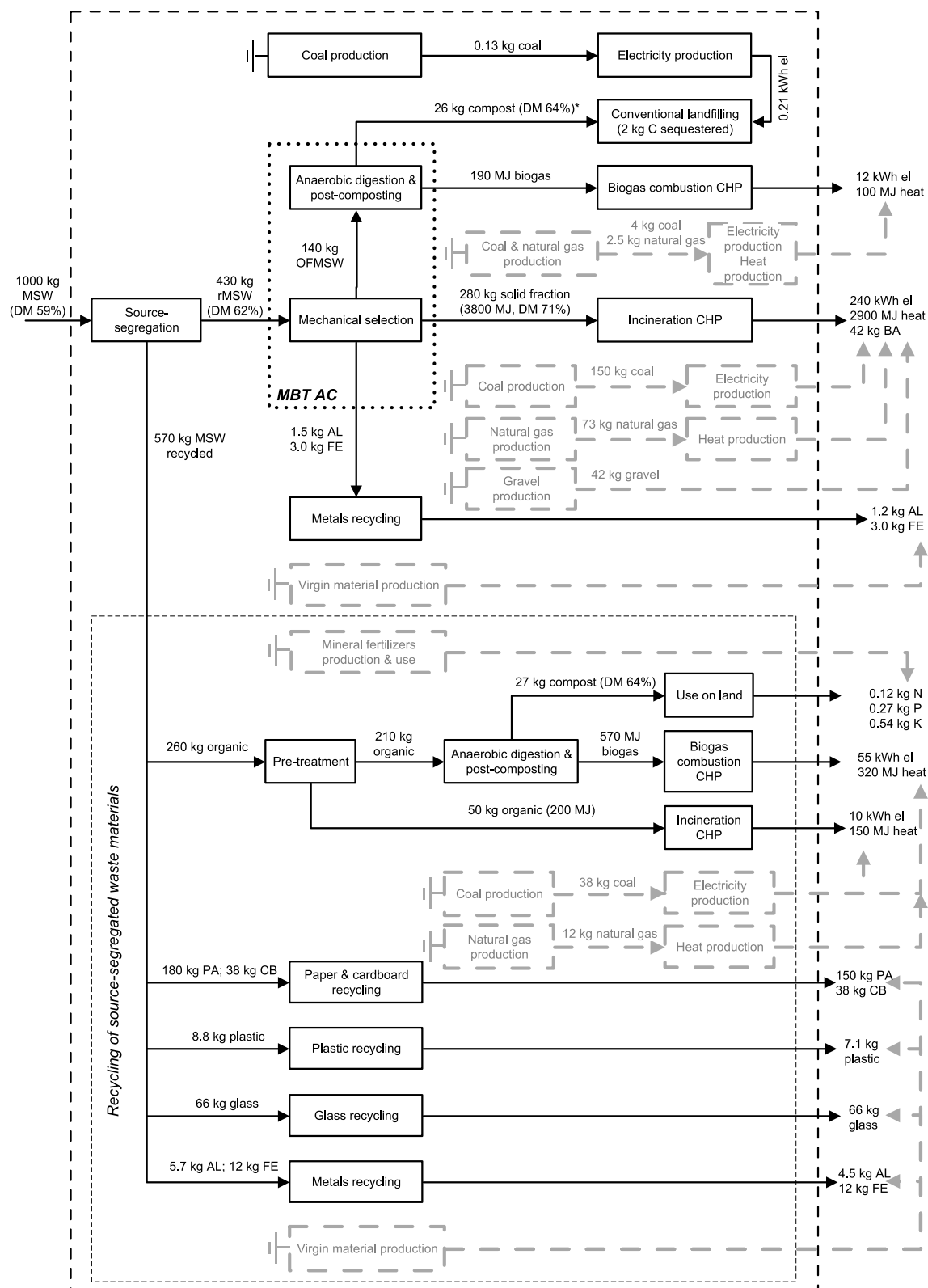


Figure S11. LCA system boundary of MBT AC I for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

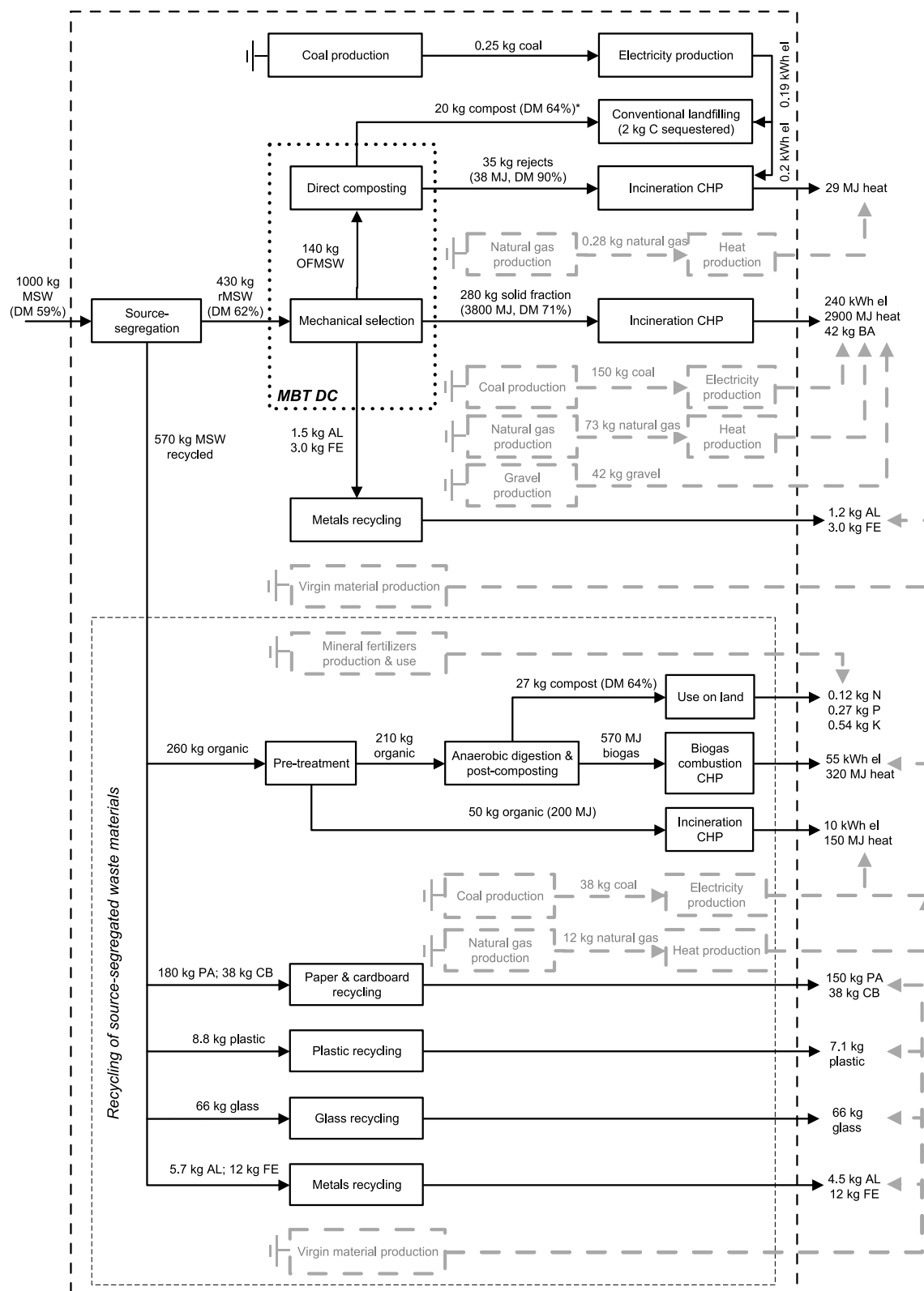


Figure S12. LCA system boundary of MBT DC I for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

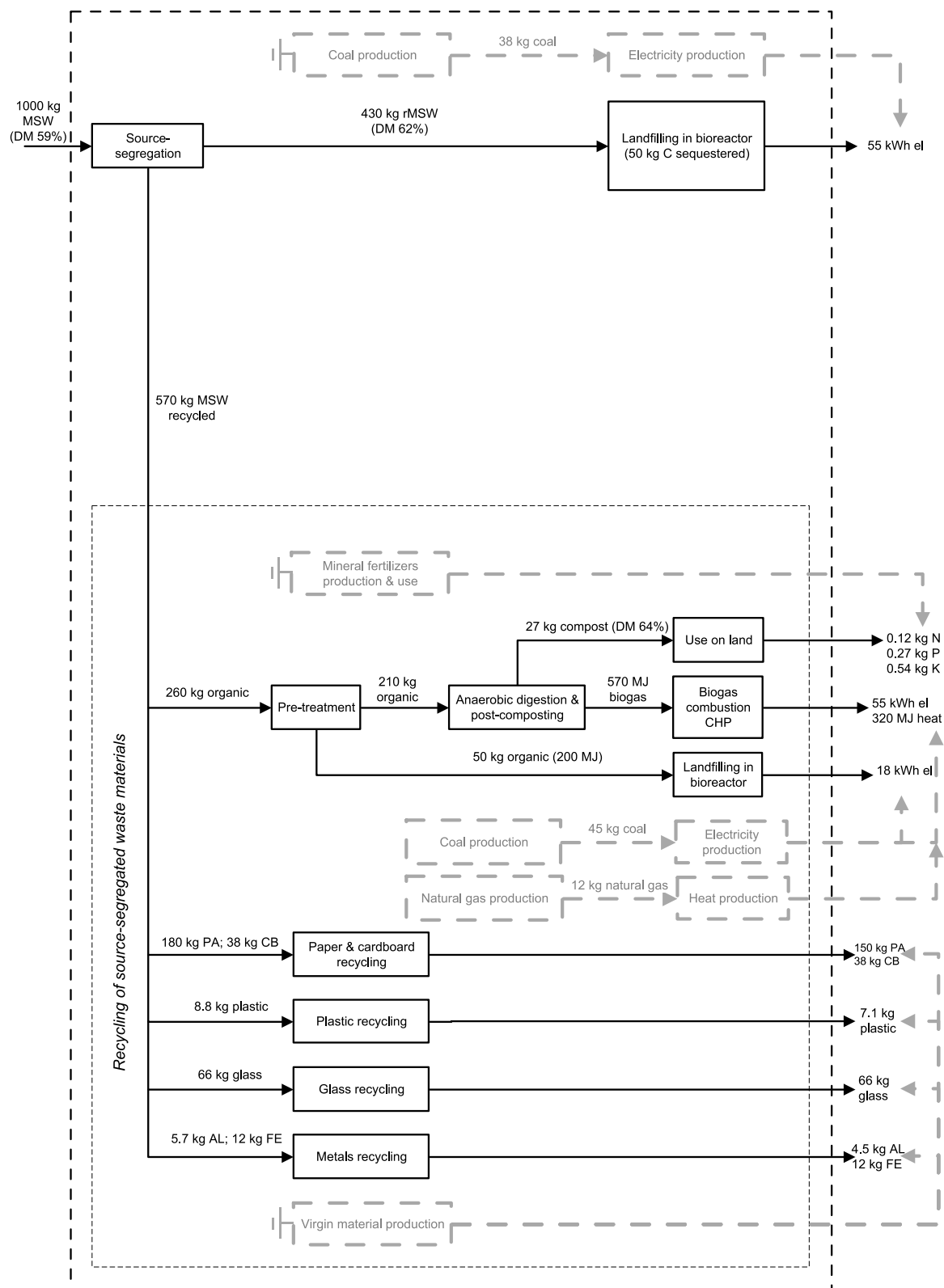


Figure S13. LCA system boundary of BLF I for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

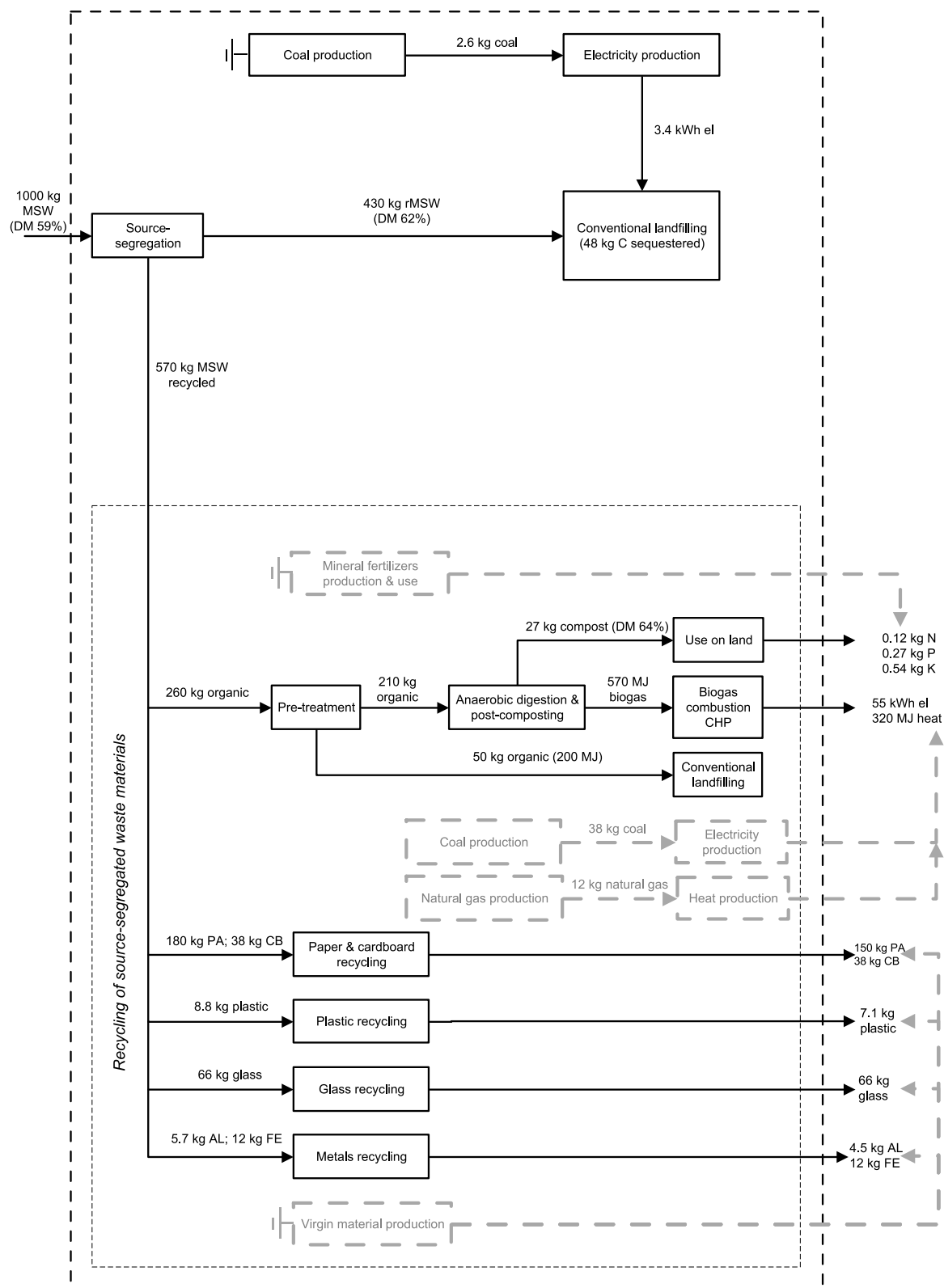


Figure S14. LCA system boundary of CLF I for the case of waste composition (a). AL: aluminium; CB: cardboard; CHP: combined-heat-and-power; FE: ferrous metal; PA: paper; rMSW: residual MSW.

Table S3. Overview of the waste management scenarios considered in the LCA. Each individual scenario was assessed with two different types of waste composition, namely (a) and (b). AD: anaerobic digestion; CHP: combined-heat-and-power; El: electricity; Enz: enzymatic; GE: natural gas engine; LF: landfill; Mec: mechanical; MSW: municipal solid waste; OFMSW: organic fraction of MSW; Res. solids: Residual solids; rMSW: residual MSW; SRF: Solid recovered fuel; SS OW: source-segregated organic waste.

Scenario		Source-segregation		Pre-treatment ^a			Associated energy conversion technologies (thermal/biological)			
		Selected materials	Organic waste	Mec	Enz	Outputs	SS OW	OFMSW/Bioliquid	SRF/Res. solids	rMSW/rejects
WR GE	0	x		x	x	Bioliquid/Res. solids		AD & GE (CHP)	Incineration (CHP)	Incineration (CHP)
WR TF		x		x	x	Bioliquid/Res. solids		AD & use for transport	Incineration (CHP)	Incineration (CHP)
INC		x								Incineration (CHP)
MBT AC		x		x		OFMSW/SRF		AD & GE (CHP)	Incineration (CHP)	Incineration (CHP)
MBT DC		x		x		OFMSW/SRF		Composting	Incineration (CHP)	Incineration (CHP)
BLF		x								Bioreactor LF (El)
CLF		x								Flaring
WR GE	I	x	x	x	x	Bioliquid/Res. solids	AD & GE (CHP)	AD & GE (CHP)	Incineration (CHP)	Incineration (CHP)
WR TF		x	x	x	x	Bioliquid/Res. solids	AD & GE (CHP)	AD & use for transport	Incineration (CHP)	Incineration (CHP)
INC		x	x	x			AD & GE (CHP)			Incineration (CHP)
MBT AC		x	x	x		OFMSW/SRF	AD & GE (CHP)	AD & GE (CHP)	Incineration (CHP)	Incineration (CHP)
MBT DC		x	x	x		OFMSW/SRF	AD & GE (CHP)	Composting	Incineration (CHP)	Incineration (CHP)
BLF		x	x	x			AD & GE (CHP)			Bioreactor LF (El)
CLF		x	x	x			AD & GE (CHP)			Flaring
WR GE	II	x	x	x	x	Bioliquid/Res. solids	AD & GE (CHP)	AD & GE (CHP)	Incineration (CHP)	Incineration (CHP)
WR TF		x	x	x	x	Bioliquid/Res. solids	AD & GE (CHP)	AD & use for transport	Incineration (CHP)	Incineration (CHP)
INC		x	x	x			AD & GE (CHP)			Incineration (CHP)
MBT AC		x	x	x		OFMSW/SRF	AD & GE (CHP)	AD & GE (CHP)	Incineration (CHP)	Incineration (CHP)
MBT DC		x	x	x		OFMSW/SRF	AD & GE (CHP)	Composting	Incineration (CHP)	Incineration (CHP)
BLF		x	x	x			AD & GE (CHP)			Bioreactor LF (El)
CLF		x	x	x			AD & GE (CHP)			Flaring

^a Intended as prior to biological or thermal energy conversion (which could be, for example, anaerobic digestion or incineration).

2. Identification of marginals

2.1 Marginal energy technologies

Special attention was devoted to assumptions regarding the surrounding energy system as choices here may significantly affect the outcome of the LCA (3-7). In a long term perspective (e.g., beyond 15 years), it may be assumed that energy from waste contributes to the decommissioning of fossil-based energy production capacities (both electricity and heat) as these technologies are generally intended to be phased out in order to comply with political CO₂ reduction targets. The waste management scenarios were therefore credited with the environmental savings induced by substitution of fossil fuel-based energy production; such system boundary expansion to include the benefits deriving from replacement of fossil energy represents a typical approach in consequential LCA (e.g., (5-7) among the others). At a European level, coal and natural gas represent the two ends of the range with respect to CO₂ emissions per combustion unit of fossil fuel energy (ca. 95 kg CO₂ GJ⁻¹ coal and 55 kg CO₂ GJ⁻¹ natural gas). These are also expected to be the fuels reacting to increased electricity production from waste and biomass. For example, in the case of Denmark (7, 8) identified coal as marginal electricity source; in the case of Italy, (9) identified natural gas as marginal. In (10) natural gas was identified as marginal for Spain. In this study it was assumed substitution of coal-based electricity production for the baseline calculation. This assumption was tested in the sensitivity analysis by substituting natural gas-based electricity production (S1).

As opposed to electricity, the market for heat is rather local and substitution of district heating or heating fuels often depends on local conditions and production capacities connected to the district heating network in question (4). This means that when evaluating a system in a short-term perspective involving existing production capacities, substitution of district heating should reflect local conditions. However, it is viable to assume that in the long-term heat production from waste will contribute to phasing-out fossil fuels. In the European Union, the heat market for Western (Austria, Denmark, Finland, France, Germany, Italy, Netherlands, Norway, Sweden, Switzerland) and Southern Europe (France, Greece, Italy, Portugal, Spain) is dominated by natural gas (ca. 45% in Western and 61% in Southern EU, relative to the total heat supply) (11, 12). In Eastern Europe (Czech Republic, Hungary, Slovak Republic, Poland) instead, the largest share of the heat supply is attributed to hard coal (ca. 55% of the total supply), followed by natural gas (ca. 22%) (11, 12). In this study, 3

possible scenarios of heat substitution were assessed to exemplify three potential (and most likely) situations that could occur in Europe (and elsewhere): i) heat produced from natural gas boiler (most representative for Western and Southern Europe), ii) heat produced from coal boiler (most representative for Eastern Europe), and iii) heat produced from coal-fired CHP plants (as an example of heat produced from CHP plants connected to an existing district heating network). District heating currently owns a low share of the EU heat market (ca. 6%). However, in the future, expansion of district heating is expected (13). In addition, most Nordic Countries have a well developed district heating network. This is, for example, the case for Denmark where about 62% of the total heat supply is provided as district heating (of this about 76% is delivered by CHP plants which main purpose is the production of electricity with heat being a co-product) (14). The marginal heat production from CHP plants was calculated following the approach of (4). Based on this, the total emissions from a representative Danish coal-fired CHP plants were allocated to electricity and heat based on the exergy content. This approach determines that ca. 90% of the emissions are attributed to electricity (main product) and the remaining to heat. The background LCI data for the marginal heat and electricity processes used in this study are reported in Table S4. The baseline results presented in the main manuscript consider substitution of heat produced from natural gas boilers. The results for the substitution of heat from coal boilers (namely S2) and of (coal-based) district heating (namely S3) are presented in Figure 2 of the manuscript. Also, an additional sensitivity analysis (namely S4) was performed to evaluate the environmental performance of the scenarios in the case of no-heat recovery.

For the transport fuel scenario (scenario WR TF), gasoline was assumed as marginal. This choice was tested in the sensitivity analysis (namely S5) by substituting diesel fuel.

Table S4. Background LCI emission data for marginal electricity and heat production used in this study (unit kg kWh⁻¹_{el} or kg kWh⁻¹_{heat}). Only selected emission data are reported.

Parameter	Coal electricity ^α	Natural gas electricity ^β	Natural gas boiler ^γ	Coal boiler ^δ	Coal District heating ^α
As	6.2E-09	3.3E-09	-	1.1E-07	1.9E-10
CO	2.1E-04	3.7E-05	1.2E-04	4.5E-04	1.7E-05
Cd	8.2E-10	6.1E-10	1.5E-10	7.1E-09	2.1E-11
CO ₂	0.91	0.45	0.22	0.41	0.17
Cu	8E-09	6.3E-09	2.4E-10	2.4E-10	2.4E-10
Cr	6.4E-09	9.6E-09	5.8E-10	1E-10	5.8E-10
Dioxins	2.1E-16	1E-016	1E-016	7.2E-014	-
Methane	5.3E-03	1.2E-05	7.8E-06	4.5E-05	3.4E-07
Hg	8.9E-09	6.1E-10	1.2E-10	1.3E-08	1.2E-09
NMVOC	8.5E-10	1.2E-05	1E-05	7.7E-06	3.4E-07
Ni	2.9E-08	1.6E-06	3.9E-12	8.5E-08	6.7E-10
N ₂ O	8.9E-6	7.7E-06	2E-06	4.5E-06	1.4E-06
NO _x	1.3E-03	3.7E-04	7.9E-05	0.0009	3.7E-04
Pb	9.2E-09	6.3E-09	2.3E-011	3.41E-10	4.2E-010
SO ₂	6.9E-04	4.3E-06	2E-06	2.3E-03	1.5E-05
Unspecified particles	1.4E-06	7.0E-07	7.2E-07	2.3E-04	6.0E-06

^α Details on data and technologies can be found in (15).

^β Based on a Danish natural gas-fired CHP plant (16).

^γ Natural gas boiler with low NO_x emissions (from Ecoinvent v2.2).

^δ A process for coal combustion in industrial furnaces was used as proxy (from Ecoinvent v2.2).

2.2 Marginal mineral N, P, and K fertilizers

The compost produced from biological treatment of source-segregated organic waste and bioliquid was used as a fertilizer (for N, P, and K), which avoided marginal mineral N, P, and K fertilizers to be produced and used, based on the content of N, P, and K of the compost. The marginal N, P, and K fertilizers considered were calcium ammonium nitrate, diammonium phosphate, and potassium chloride, respectively, conformingly with (17, 18). Based on these, calcium ammonium nitrate is the N-fertilizer that is most likely to react to additional use of N organic fertilizer in a EU perspective. Diammonium phosphate and potassium chloride are those, in a EU and World perspective, expected to react to an additional use of P and K organic fertilizer, respectively: the first because current market trends show that new investments on P-fertilizers producing capacities are mainly for diammonium phosphate production, and the second because it currently accounts for ca. 95% of the all K fertilizers used in agriculture (19).

3. LCI of waste treatment technologies

This section describes the waste treatment technologies used for the assessment of the 252 waste management scenarios. Relevant technologies efficiencies have been forecasted to consider future development and optimization: this was done with respect to energy efficiency, selected air emissions, leachate and gas treatment at the landfill, use of enzymes and energy at the waste refinery, and biogas yield. Table S7 provides an overview of energy and material input and output to and from the technologies used in the assessment. Table S8 highlights future technologies development (as modelled in this study) compared with current typical operational efficiencies. Data are based on a number of literature sources.

3.1 Pre-treatments

This section describes the waste pre-treatment technologies used in the assessment: i) waste refining, ii) mechanical-biological treatment (MBT), and iii) mechanical selection of source-segregated organic waste prior to biological treatment.

3.1.1 Waste refinery

The waste refinery process was based on a pilot-scale facility established in Copenhagen, Denmark. The waste refinery had a treatment capacity of about 1 t MSW h⁻¹ (wet weight) and has been in operation from 2010.

The waste refinery (Figure S15) aims at producing two primary products from the incoming mixed MSW: i) a bioliquid (i.e., slurry composed of enzymatically liquefied organic, paper, and cardboard) and ii) a residual solid (i.e., non-degradable waste materials). The refinery process consisted of two reactors: in the first reactor the waste was heated with hot water to about 75 °C for approximately 0.5-1 hours, then cooled to about 50-55 °C before entering the second reactor. In the second reactor enzymes were added (about 5 kg t⁻¹ MSW) resulting in hydrolysis and break-down of bonds in the organic materials thereby essentially suspending organic materials in a liquid phase. The retention time was about 10-16 hour. A detailed description of the enzymatic processing can be found in (20). After the second reactor, the liquid phase was separated from the remaining solids by a vibrating sieve. Further, another vibrating sieve separated the liquid phase into a bioliquid and a solid “fluff” (phase containing materials such as cotton and textiles, but also glass pieces, plastics, etc.).

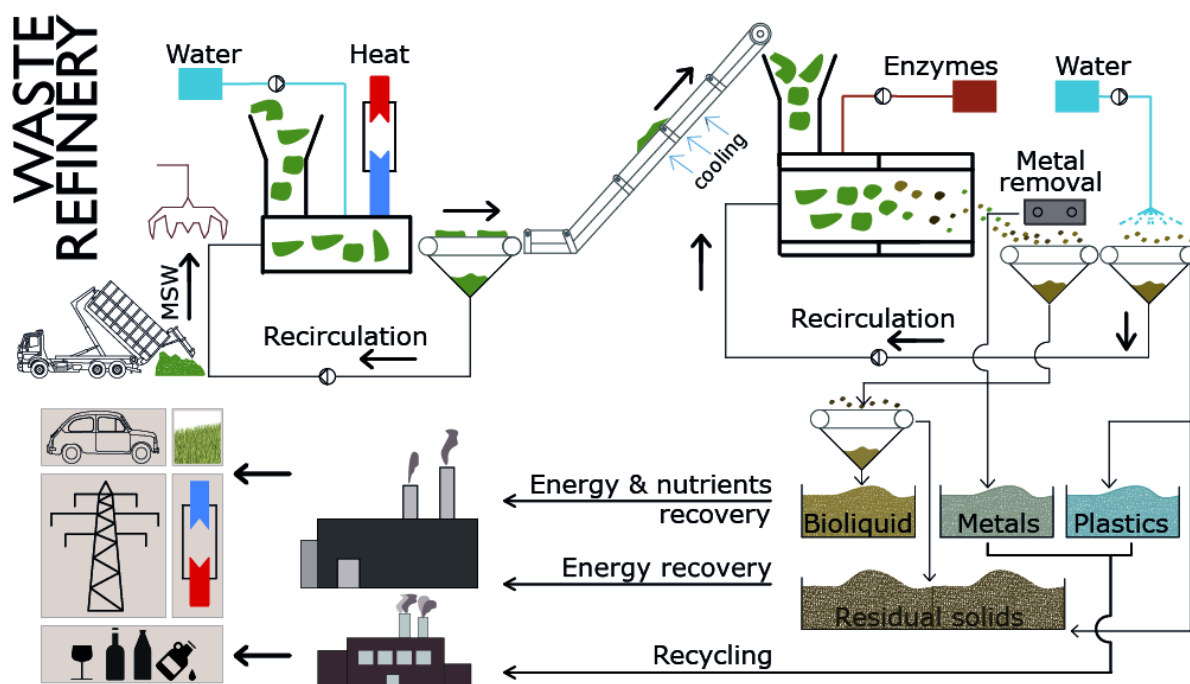


Figure S15. Illustration of the waste refinery process.

The bioliquid consisted primarily of suspended organic matter (food waste, paper, and cardboard), while the solid fraction mainly consisted of non-degradable materials such as plastic, metals, textiles, soil, ceramics, etc. The bioliquid can be exploited for biogas production (option considered in this study and described in section 3.3 dedicated to biological treatments), co-combusted in coal-fired power plant or utilized for producing ethanol. This, compared with direct incineration, provides additional flexibility to the energy system as the energy production could be regulated and storage possible in form of bioliquid/biogas. This is important in the perspective of energy systems having high penetration of wind energy and other fluctuating renewables as illustrated in previous studies (21-25). The solid fraction can be further treated to separate and recover valuable materials such as metals and (eventually, if applied) plastic. The remaining residual solids (mainly non-recyclable plastic, textiles, yard waste, undegraded organics, glass pieces, and fluff) can be combusted for energy recovery. The amount of bioliquid and residual solids fraction produced in the process vary upon the composition of the incoming waste. To this regard, a list of transfer coefficients used to model the materials transfer to bioliquid and residual solid fraction can be found in Table S5; these values were experimentally determined (after (20)). Table S6 reports average chemical composition data for the bioliquid as experimentally determined. Overall, the electricity and heat consumption for the operations of heating, enzymatic treatment, and sieving was about 20 kWh t⁻¹ MSW and 490 MJ t⁻¹ MSW. These

were based on process data from the operations of the pilot-scale plant. For metals separation additional 4.5 kWh t^{-1} MSW (magnet and ECS) were considered based on the operational data for mechanical recovery facilities provided in (26). In the case that also plastic was separated (HDPE, HDPP, and PET), additional 8.2 kWh t^{-1} MSW should be considered (26) (optical separation of PET and HDPE, plus baling). In this study plastic recovery was not considered as previous results showed negligible environmental differences between recovering it and not (27). Additional information about the process and its environmental performance relative to a Danish incinerator can be found in (27).

Table S5. Bioliquid from waste refining: transfer coefficients of selected waste material fractions. After (20).

Waste material fraction	TC (% of input)
Vegetable and animal waste	100%
Paper and cardboard	100%
Cartons and alike	85%
Animals litter	50%
Soil/ash	50%
Yard waste	35%
Aluminium foil	30%
Diapers/sanitary towel/tampons/bandages/cotton	15%

Table S6. Bioliquid from waste refining: chemical composition in kg kg^{-1} DM (unpublished results from analyses carried out at a Danish pilot-scale refinery). The LHV_{db} and CH_4 pot are expressed as MJ kg^{-1} DM and $\text{NL CH}_4 \text{ kg}^{-1}$ VS, respectively. Values are rounded to two significant digits.

C_{biog}	H	S	Cl	N	P	K	Fe	Al	Cd	Cr
4.2E-01	5.1E-02	1.0E-03	1.1E-02	2.0E-02	2.5E-03	8.4E-03	5.1E-03	3.0E-03	1.8E-07	1.9E-05
Cu	Ni	Sr	Mn	As	Hg	Pb	Sb	O	LHV_{db}	CH_4 pot
3.5E-05	1.2E-05	1.0E-04	8.0E-05	1.4E-06	1.0E-07	1.2E-05	1.4E-06	4.7E-01	16	440

3.1.2 Mechanical-biological treatment (MBT)

Two types of mechanical-biological plants were modelled: i) MBT AC with anaerobic digestion and post-composting of the OFMSW (organic fraction of municipal solid waste) and ii) MBT DC with direct composting of the OFMSW. These technologies (and therefore the related scenarios) differed for the biological treatment, while the mechanical pre-treatment was the same. The data were based on a full-scale MBT plant operated in Spain (28). The plant had four main outputs: ferrous metals, aluminium, OFMSW and solid

recovered fuel (SRF). The recovery of ferrous and non-ferrous was set to 85% and 90%, respectively, in order to have consistency across the assumptions about metals recovery in MBT and waste refinery, being the mechanical selection based on the same type of technology (magnets and eddy current systems, i.e., ECS). The authors are aware of that other materials (e.g., paper, plastic, and glass) could be further recovered in MBT plants; however, this was not considered in the present as the largest share of recyclables was assumed source-segregated from the MSW with high efficiency (see Figure S1-S14); this justified a ‘simpler’ MBT technology for the treatment of the rMSW where the primary concern is energy recovery and stabilization of organic waste along with production of a high-calorific value fraction (SRF) for further energy recovery in highly-efficient incineration.

The mechanical treatment consisted of shredding (bags opening), trommel for separation of the organic fraction (OFMSW), and of separation of ferrous and non-ferrous metals by means of magnets and ECS, respectively. The transfer coefficients for organic, ferrous and non-ferrous metals corresponded to 93%, 85%, and 90%. The residual solid fraction (SRF) was assumed to be incinerated for energy recovery. The consumption of electricity and diesel for the mechanical operations corresponded to 15 kWh t⁻¹ MSW and 0.32 t t⁻¹ MSW. The associated biological treatments for the mechanically separated OFMSW are detailed in section 3.3.

3.1.3 Mechanical-selection of source-segregated organic waste

Prior to anaerobic digestion, source-segregated organic waste generally needs mechanical pre-treatment in order to remove unwanted items (e.g. plastic, large wood pieces, packaging materials, and other misplaced items), to reduce particle sizes before the reactor thereby minimizing mechanical problems, to mix several substrates, to enhance hygienization, and adjust waste properties (29). This leads to losses of organic matter as a consequence of inherent efficiencies of the mechanical separation. Based on recent field investigations on a number of existing anaerobic digestion plants with associated pre-treatment (29), these losses might fall in the range 13-39% (as percent of DM of the incoming waste). Based on these results, it was assumed that 20% (as average value) of the source-segregated organic waste input to the anaerobic digestion plant was diverted to the reject fraction during the pre-treatment. This was also in accordance with the findings of (30). The reject was further routed to the ‘reference technology’ for residual waste treatment (that is, incineration for

INC, landfill for CLF and BLF, and incineration for MBT AC, MBT DC, WR GE and WR TF). Electricity and diesel fuel consumption were assumed as for mechanical selection at MBT plants (15 kWh t^{-1} OFMSW and 0.32 t t^{-1} MSW). The associated anaerobic digestion process for the pre-treated source-segregated organic waste is described in the section dedicated to biological treatments (section 3.3).

3.2 Waste incineration

The waste incineration plant was modelled as a grate-fired incinerator equipped with wet flue gas cleaning, selective catalytic reduction (SCR) of NO_x , Hg and dioxin removal by activated carbon. The SCR-process for NO_x reduction (currently not a legal requirement in many EU Countries) determines energy use for heating the flue-gas and electricity losses due to increased pressure loss. This typically causes a reduction of about 1% of electricity generated and ca. 2.5% of total energy recovered (31). Gross electricity and heat efficiencies were therefore assumed to 25% and 75%, respectively, relative to the LHV_{wb} of the waste input conformingly with expected performances for 2015-2030 (31) (see Table S8). The total gross efficiency added up to 100%, relative to the LHV_{wb} , as the plant was assumed provided with flue-gas condensation. Based on target cleaning efficiencies, the materials and resources consumption for operations and flue-gas cleaning were estimated: internal electricity consumption at the plant was 86 kWh t^{-1} MSW plus an additional 0.63 L t^{-1} MSW of oil as auxiliary fuel, $0.19 \text{ kg NaOH t}^{-1}$ MSW, $0.8 \text{ kg NH}_3 \text{ t}^{-1}$ MSW, $0.5 \text{ kg activated carbon t}^{-1}$ MSW and $11 \text{ kg CaCO}_3 \text{ t}^{-1}$ MSW for flue-gas cleaning.

Following the approach of (32), emissions were divided into either process-specific emissions (emissions independent of waste composition but proportional to the amount of waste incinerated) or waste-specific emissions (determined by output transfer coefficients). The emission of NO_x was assumed 0.13 g t^{-1} MSW (15 g GJ^{-1} waste input) as expected with SCR (typically $< 30 \text{ g GJ}^{-1}$ (31)). The SO_2 emission equalled 0.013 kg t^{-1} MSW which assumed a degree of desulphurisation higher than 98.5% (31). This is in line with typical SO_2 emissions for Danish incinerators (33). Selected air emissions for relevant heavy metals were (as % of input): 0.2% (As), 0.1% (Cd), 0.01% (Cr), 0.0018% (Cu), 0.004% (Mn), 0.125% (Ni) and 0.015% (Pb) conformingly with available operational data from a Danish incinerator (34). The distribution of Al between bottom and fly ash was ca. 90.5% and 9%, respectively. Similar transfer coefficients were reported for another Danish waste-to-energy plant (27). Notice that the transfer coefficient of Hg to air was set to 0.5% (% of Hg input): this assumes

improved removal thanks to absorption in filters as indicated in previous studies focusing on the historical development of air pollution control (35). The current performances indicate higher transfer emissions (ca. 3.5-5% of the Hg input) (27, 34).

Ferrous and non-ferrous metals were recovered from bottom ashes (BA) prior to utilization as construction material substituting natural gravel (36). The recovery efficiency was set to 85% for ferrous metals and 70% for non-ferrous (here assumed as aluminium). These values have to be considered as typical for state-of-the-art European recovery technologies based on the findings of a recent review (37). The electricity and diesel consumption for the operations equalled $1.3 \text{ kWh t}^{-1} \text{ BA}$ and $1.1 \text{ L t}^{-1} \text{ BA}$. Fly ashes were assumed to be utilized in the backfilling of old salt mines (38).

3.3 Biological treatment

3.3.1 *Biological treatment of source-segregated organic waste*

The source-segregated organic waste was digested in a one-stage mesophilic anaerobic digestion plant appropriate for OFMSW (39).

The methane yield was set to 75% of the potential in agreement with similar practice for organic waste (40-42) yielding about $290\text{-}320 \text{ Nm}^3 \text{ CH}_4 \text{ t}^{-1} \text{ VS}$ (depending on the waste composition). The methane content in the biogas was assumed to be 60% (v/v). Internal energy consumption at the plant was: diesel ($0.9 \text{ kg t}^{-1} \text{ OFMSW}$), electricity ($40 \text{ kWh t}^{-1} \text{ OFMSW}$) and heat ($100\text{-}110 \text{ MJ t}^{-1} \text{ OFMSW}$). The electricity was due to: digestion operations ($18 \text{ kWh t}^{-1} \text{ OFMSW}$ for pumping, ventilation, etc.), dewatering by pressing (about $7 \text{ kWh t}^{-1} \text{ OFMSW}$), and post-composting and sieving ($15 \text{ kWh t}^{-1} \text{ OFMSW}$). These data were based on (42, 43). The amount of heat required was calculated based on the energy required to heat up the substrate (about 35-40% DM, depending on the considered waste composition) from $8 \text{ }^\circ\text{C}$ to $37 \text{ }^\circ\text{C}$ and to maintain constant temperature ($37 \text{ }^\circ\text{C}$) in the reactor. The fugitive emissions of CH_4 were assumed 1% of the methane produced in accordance with recent LCA studies (17, 43, 44).

The biogas was assumed combusted in a natural gas engine with electricity and heat efficiency of 40% and 60% relative to the LHV of the biogas. Similarly to incineration, the total efficiency added up to 100% as the plant was assumed provided with flue-gas condensation. This was done in order to have consistency regarding the assumptions about heat recovery. In fact, although not yet established, flue-gas condensation can be applied to natural gas engines achieving energy recoveries greater than 100% (relative to the LHV of the biogas) as for incineration (31). Yet, it has to be noted that this represents an upper limit

as typically these installations have smaller capacity (5-8 MW per unit) than incinerators (typically larger than 100 MW) and this is an inherent limitation for the implementation of expensive flue-gas condensation systems. Air emissions from biogas combustion in natural gas engines were assumed according to (33).

The digestate was post-composted in aerated tunnels provided with biofilters for exhaust gas cleaning; the degradation of the organic matter during post-composting was assumed between 15% (organic waste, e.g., vegetable, animal waste, animals litter, etc.) and 20% (paper, cardboard, cotton, textiles, wood, etc.). The fugitive emissions of CH₄ occurring during the process were assumed 0.2% of the C degraded, and N₂O emissions to 1.4% of the N degraded. About 98.5% of the degraded N was in the form of NH₃ of which 99% was oxidized in biofilters in line with (39) for a similar composting technology. A final sieving of the unrefined compost was also modelled; this implied that unwanted materials such as diapers, cotton, wood, textiles and undegraded paper were rejected and sent to residual waste treatment (see Figures S1-S14). This constituted ca. 5% of the unrefined compost mass. The DM content of the refined compost was set to 64% conformingly with (39).

3.3.2 Biological treatment of OFMSW (MBT with anaerobic digestion and post-composting)

For MBT with anaerobic digestion and post-composting (MBT AC) anaerobic digestion of the OFMSW was modelled based on the data for a MBT plant operating in Spain (28). Accordingly, the methane yield was 55% of the potential of the incoming waste. The methane content in the biogas was 60% (v/v). The fugitive methane emissions from the reactor were set to 1% of the produced CH₄ based on recent LCA studies (17, 43, 44). The energy consumption was calculated as for source-segregated organic waste digestion and corresponded to: electricity (25 kWh t⁻¹ OFMSW for digestion operations and dewatering), heat (100-110 MJ t⁻¹ OFMSW, depending on water content) and diesel (0.9 kg t⁻¹ OFMSW).

The biogas was assumed combusted in a natural gas engine with electricity and heat efficiency of 40% and 60% relative to the LHV of the biogas. Similarly to incineration, the total efficiency added up to 100% as the plant was provided with flue-gas condensation. Air emissions from biogas combustion in natural gas engines were assumed according to (33).

After 15-20 days of digestion, the digestate was introduced to post-composting. This consisted of forced aeration in tunnels (15-20 days) followed by maturation in piles (about 2 months). The degradation of each material fraction was modelled as a percentage of the VS content of the incoming waste; this corresponded to about 35-40% for paper and organic materials, respectively. The electricity consumption was estimated to 15 kWh t⁻¹ OFMSW

(adapted from (39)). The air emissions were modelled as for MBT DC (see later description). The rejects from the process were assumed incinerated. The compost was assumed landfilled according to current practices.

3.3.3 Biological treatment of OFMSW (MBT with direct composting)

For MBT with direct composting of the OFMSW (MBT DC), the biological treatment was modelled as aerated tunnels provided with biofilters for exhaust gas cleaning conformingly with (28). The degradation of each material fraction was modelled as a percent of the VS content in the incoming waste; this corresponded to about 70% VS degradation for organic waste such as animal and vegetable food waste, 60% for yard waste, 10-20% for paper and cardboard fractions, and 5% for beverage cartons and textiles. The electricity consumption was 40 kWh t⁻¹ OFMSW similarly to source-segregated organic waste. The main emissions were CH₄, N₂O, and NH₃. Conformingly with (28, 39), the fugitive CH₄ emissions were set to 0.2% of the degraded C, and N₂O emissions to 1.4% of the degraded N. About 98.5% of the degraded N was in the form of NH₃ of which 99% was oxidized in biofilters (39) which are the current air treatment system at the considered plants. The rejects from the process were assumed incinerated. The compost was assumed landfilled according to current practices.

3.3.4 Biological treatment of the bioliquid from waste refinery

For bioliquid from waste refining, the methane yield was set to 82% of the methane potential based on the results of tests conducted in a full-scale biogas plant (unpublished results) yielding about 365 Nm³ CH₄ t⁻¹ VS. The methane content in the biogas was assumed to be 60% (v/v). Internal energy consumption at the plant was: diesel (0.9 kg t⁻¹ bioliquid), electricity (40 kWh t⁻¹ bioliquid) and heat (ca. 120 MJ t⁻¹ bioliquid). The amount of heat required was calculated based on the energy required to heat up the substrate (20% DM) from 8 °C to 37 °C. The fugitive emissions of CH₄ were assumed 1% of the methane produced in accordance with recent LCA studies (17, 43, 44).

The biogas was assumed combusted in a natural gas engine with electricity and heat efficiency of 40% and 60% relative to the LHV of the biogas. Similarly to incineration, the total efficiency added up to 100% as the plant was assumed provided with flue-gas condensation. Air emissions from biogas combustion in natural gas engines were assumed according to (33).

Similarly to the treatment of source-segregated organic waste, the digestate was post-composted in aerated tunnels provided with biofilters for exhaust gas cleaning. Emissions and

process efficiencies were assumed accordingly, except that no rejects were considered from the process.

3.4 Landfilling in bioreactor

The inputs to the bioreactor landfill were electricity ($10 \text{ kWh t}^{-1} \text{ MSW}$), diesel oil for vehicles operating on site ($2 \text{ L t}^{-1} \text{ MSW}$), soil and clay (in total, $0.5 \text{ t t}^{-1} \text{ MSW}$). The soil and clay were assumed to be transported 30 km on average. The landfill had an average filling depth of 20 m and a bulk waste density of $1.2 \text{ t m}^{-3} \text{ MSW}$.

The generated leachate was recirculated to the waste mass for a time period of 8 years, in order to optimize the waste degradation process and increase the rate of methane generation. Tap water was also added in order to achieve an amount of recirculated water equal to 6750 mm within 8 years. During this period, 65% of the overall methane potential was generated and 90% of this collected. The collected gas was extensively (90%) used for electricity production in a natural gas engine with an efficiency of 40% relative to the LHV of the biogas. Gas collection continued for 10 years after the end of the leachate recirculation period. During this period, 20% of the overall methane potential was generated and 90% of this collected. The collected gas was used (70%) for electricity production with the same efficiency as earlier. The collected gas which was not used for electricity generation was instead flared. The uncollected gas fraction received partial oxidation in the top soil cover. Oxidation efficiencies were specified for each gas constituent for all time periods. With respect to methane oxidation, efficiencies ranged from 40% to 80% depending on the actual flow-rate of gas through the cover: 40% was used until the end of the leachate recirculation period and 80% oxidation afterwards.

During the first 2 years of filling operations the leachate generation was set to 450 mm y^{-1} . After the first 2 years an impermeable surface liner was applied as top cover. During the following 13 years the leachate generation in the model was set to 450 mm y^{-1} : in reality this is equivalent to 8 years of leachate recirculation (6,750 mm including household water addition) followed by 5 years of draining operations. This way of modeling did not reflect how the water actually came out (this is not really important as the emissions in the modelling are integrated over 100 years) but it reflected that all the added water was supposed to come out before 15 years after the start of the landfill. After the end of the draining period, the leachate generation was set to 20 mm y^{-1} , due to the impermeable top sealing. The leachate collection system was assumed to capture 95% of the leachate generated for the first 40 years of the landfill life time. Thereafter, since only 20 mm y^{-1} of

leachate was generated, the leachate collection efficiency was set to 0% and all the uncollected leachate migrated to the groundwater. The collected leachate was treated in a leachate treatment plant for purification. Treated leachate was discharged to surface water bodies. Natural attenuation of leachate moving towards the groundwater has been disregarded for all leachate constituents except ammonia. It was assumed that half of the ammonia was converted into nitrates, as the redox conditions in the subsurface may be favourable to oxidation.

3.5 Conventional landfilling

Conventional landfilling was modelled as an up-to-date conventional landfill with engineered measures to prevent emissions of gas and leachate to the environment. These included bottom liner, leachate collection system and leachate treatment prior to discharge of treated leachate to surface water bodies, and top soil cover, gas collection system and flaring. Also the provision of energy and materials to site and on-site operations were included. Data were adapted from (45).

The inputs to the landfill were electricity ($8 \text{ kWh t}^{-1} \text{ MSW}$), diesel oil for vehicles operating on site ($2 \text{ L t}^{-1} \text{ MSW}$), soil and clay (in total, $0.5 \text{ t t}^{-1} \text{ MSW}$). The soil and clay were assumed to be transported 30 km on average. The landfill had an average filling depth of 20 m and a bulk waste density of $1 \text{ t m}^{-3} \text{ MSW}$.

The filling phase was divided into two parts. In the first part (years 1-2) no gas collection was practiced, while in the second part (years 3-5) gas was extracted. It has been assumed that gas collection and gas treatment was active until year 100. Gas generation and collection were modelled according to 4 time periods: during the first 2 years of sector filling, 1% of the overall methane potential was generated and no-collection was practiced. During the following 3 years of sector filling, 4% of the overall methane potential was generated and 50% of this was collected. This inherently assumed that from year-3 gas wells were constructed and operated as the landfill body grew higher. The final soil top cover was constructed at the end of the filling phase (year-5). The final cover did not include a surface liner. However, assuming a soil cover only is in compliance with EU regulations and represents a worst-case scenario allowing for leachate generation and migration of LFG through the cover. Within the following 25 years, 60% of the overall methane potential was generated and 50% of this was collected. During the remaining 70 years, 30% of the overall methane potential was generated and again 50% of this was collected. All the collected gas during 100 years was flared. The uncollected gas fraction received partial oxidation in the top

soil cover. Oxidation efficiencies were specified for each gas constituent for all time periods. With respect to methane oxidation, efficiencies ranging from 40% to 80% were assumed depending on the actual flow-rate of gas through the cover.

During the first 2 years of filling operations the leachate generation was set to 350 mm y⁻¹. In the following 3 years of filling the leachate generation was set to 300 mm y⁻¹, assuming major water retention due to the increased waste mass. After the soil top cover construction (year-5) the leachate generation dropped to 200 mm y⁻¹. Afterward, from year 30 to year 100 the leachate generation was decreased to 100 mm y⁻¹, assuming an increased evapotranspiration due to vegetation growth on the top cover. Leachate collection and treatment were assumed operated until year 30. With respect to leachate collection, capture efficiencies were set to 95% during the first 30 years of the landfill life. Afterwards (from year 30 to year 100), an impermeable surface liner was constructed and the leachate collection stopped. The capture efficiency was then set to 0% and all the leachate generated was assumed to reach the groundwater.

The collected leachate was treated in a leachate treatment plant for purification. Treated leachate was discharged to surface water bodies. Natural attenuation of leachate moving towards the groundwater has been disregarded for all leachate constituents except ammonia. It was assumed that half of the ammonia is converted into nitrates, as the redox conditions in the subsurface may become favourable to oxidation.

Table S7 Overview of energy and materials input and output to and from the main technologies used in the LCA. El: electricity; APC: air pollution control residue; MSW: municipal solid waste; OFMSW: organic fraction of MSW; Res. solids: Residual solids; rMSW: residual MSW; SRF: Solid recovered fuel; SS OW: source-segregated organic waste.

Technology		Input				Output					
		Materials		Energy		Materials/products			Energy		
		Waste materials	El (kWh t ⁻¹)	Heat (MJ t ⁻¹)	Fuel (L t ⁻¹)	Ferrous (% input)	Aluminium (% input)	Other outputs	El ^β (%)	Heat ^β (%)	Technology
Pre-treatment	Mechanical selection	rMSW, SS OW	15	-	0.32	85	90	OFMSW SRF	-	-	-
	Waste refining	rMSW	25-33 ^a	490	-	85	90	Bioliquid Res. solids	-	-	-
Biological treatment	Anaerobic digestion of SS OW from source-segregation	SS OW	40	100-110	0.9	-	-	Biogas Compost	40	60 ^γ	Natural gas engine (CHP)
	Anaerobic digestion of OFMSW from MBT	OFMSW	40	100-110	0.9	-	-	Biogas Compost	40	60 ^γ	Natural gas engine (CHP)
	Direct composting of OFMSW from MBT	OFMSW	40	-	0.9	-	-	Compost	-	-	-
	Anaerobic digestion of bioliquid from waste refinery	Bioliquid	40	120	0.9	-	-	Biogas Compost	40	60 ^γ	Natural gas engine (CHP)
rMSW treatment	Incineration	rMSW, SRF, Res. solids	86	-	0.63 ^δ	85	70	Bottom ash APC	25	75 ^γ	Steam cycle (CHP)
	Conventional landfilling	rMSW	8	-	2	-	-	Biogas Leachate	-	-	-
	Bioreactor landfilling	rMSW	10	-	2	-	-	Biogas Leachate	40	-	Natural gas engine (El)

^α The value 33 kWh includes plastic separation (ca. 8.2 kWh). ^β Gross efficiency, expressed as percent of LHV_{wb} of the incoming waste (except for biogas for which the energy content is expressed as LHV_{db}). ^γ Flue-gas condensation is assumed (total energy recovery 100%; thermal energy losses due to SCR are accounted for). ^δ Auxiliary fuel (fuel oil).

Table S8. Overview of future expected performances of the technologies used in the assessment (selected efficiencies and/or operational data). For the purpose of comparison, current (typical) gross efficiencies and/or operational data are reported based on different sources. The reported total efficiencies are to be considered at full-load (i.e., dismissing of surplus heat in cooling towers during summer is not considered).

Technology	Current (typical) performance (2012)	Expected future performance (2015-2030)	Note
Mechanical selection ^α	Ferrous metal recovery: 65-90% (46) Aluminium recovery: 60-90% (46)	Ferrous metal recovery: 85% Aluminium recovery: 90%	Optimized material recovery.
Waste refining	El consumption: 33-51 kWh t ⁻¹ (27) Heat consumption: 590 MJ t ⁻¹ (27) Enzymes consumption: 12-24 kg t ⁻¹ (27)	El consumption: 25-31 kWh t ⁻¹ Heat consumption: 490 MJ t ⁻¹ Enzymes consumption: 5 kg t ⁻¹	Optimized energy and enzyme consumption (updated data from pilot-scale plant).
Biological treatment (anaerobic digestion and biogas combustion in a natural gas engine CHP)	Air emissions: CH ₄ (fugitive): 0-3% of CH ₄ produced (47, 48) CH ₄ yield: 60-90% of potential (49) El efficiency: 34-42% (50) Total efficiency: 85-90% (31)	Air emissions: CH ₄ (fugitive): 1% of CH ₄ produced (17, 43, 44) CH ₄ yield: 75% of potential (40-42) El efficiency: 40-50% (31) Total efficiency: 88-100% (31)	Improved insulation of the reactor. Optimized energy recovery by means of flue-gas condensation.
Biological treatment (direct and post-composting)	Air emissions: CH ₄ : 0.8-13.5% of C degraded (51) ^β N ₂ O: 0.1-1.8% of input N (51) ^β	Air emissions: CH ₄ : 0.2% of C degraded (39) N ₂ O: 1.4% of input N (39)	Post-composting is operated in aerated tunnels instead of outdoor piles. This allows for reducing CH ₄ emissions as the exhaust air is oxidized in biofilters.
Incineration	El consumption: 67 kWh t ⁻¹ (34) El efficiency: 14-21% (52) Total efficiency ^θ : up to 102% (31) Air emissions: Hg: 3.5-5% of Hg input (27, 34) NO _x : 80-180 g GJ ⁻¹ (input fuel) (31)	El consumption: 86 kWh t ⁻¹ El efficiency: 25-30% (31) Total efficiency ^γ : 100% (31) Air emissions: Hg: 0.5% of Hg input (35) NO _x : < 30 g GJ ⁻¹ (input fuel) (31) ^γ	Optimized technology equipped with SCR, activated carbon system, and flue-gas condensation. This allows for increased efficiency for energy recovery and NO _x and Hg removal.
Bioreactor landfilling	-	See section 3.4 (45)	State-of-the-art bioreactor landfill
Conventional landfilling	-	See section 3.5 (45)	State-of-the-art conventional landfill

^α Mechanical selection is used in MBT prior to biological treatment and as pre-treatment for source-segregated organic waste prior to digestion. However, in the latter case ferrous metal and aluminium recovery were not modelled as these are typically disposed of in landfill since the materials are very dirty (53). ^β Review of a number of different studies.

^γ From 2020 application of the SCR-process (selective catalytic reduction) is assumed for NO_x reduction (currently not a legal requirement in many EU Countries). Energy use for heating the flue-gas and electricity losses due to increased pressure loss typically causes a reduction of about 1% of electricity generated and ca. 2.5% of total energy recovered (included in the forecasted values for those parameters). ^θ Depends upon heat demand and presence of a district heating network (the value 102% represents a potential maximum total recovery with flue-gas cleaning and with SNCR for NO_x reduction for Danish conditions according to (31)).

3.6 Recycling

Ferrous metal recycling was assumed to substitute 98% virgin production (i.e., 2% material loss) with a market substitution ratio of 100% for the produced metal. Ferrous metal recycling included re-melting of scraps and rolling of new steel sheets from the melted metal waste, minus the avoided virgin production. The benefit of metal recycling was primarily savings in energy consumption, corresponding to a net saving of 1,700 kg CO₂-eq. t⁻¹ ferrous metal input (when coal was the marginal energy in the recycling process).

Aluminium recycling was assumed to substitute 79% virgin production (i.e., 21% material loss) with a market substitution ratio of 100% for the produced aluminium. An overall material loss of 21% was assumed due to the sorting process. Aluminium recycling included re-melting of aluminium scrap and alloying, minus the avoided virgin production. The benefit of aluminium recycling was primarily savings in energy consumption, corresponding to a net saving of 7,700 kg CO₂-eq. t⁻¹ aluminium input (when coal was the marginal energy in the recycling process).

Plastic recycling was assumed to substitute 90% virgin production (i.e., 10% material loss) by re-melting with a market substitution ratio of 90%. The latter value was a rough estimate of the potential decrease in material quality. According to (54), the loss of material quality can be as high as 20%. However, in (54) it is also stated that the loss highly depends on the field of application of the secondary plastic. For instance, in the case that the recovered plastic is utilized as an admixture in the production of primary plastic there may be no loss. Thus, in this study, 10% loss in material quality was assumed. Plastic recycling included the granulation and re-melting for production of polyethylene (PE) plastic from waste plastic minus the avoided virgin production. The benefit of plastic recycling was primarily savings in energy consumption, corresponding to a net saving of 810 kg CO₂-eq. t⁻¹ plastic input (when coal was the marginal energy in the recycling process).

The recycling dataset used for paper reflected European paper recycling processes around year 2000, although not necessarily statistically based. Newspaper and magazines were recycled into newspaper. The technical substitution was 82% (i.e., 18% material loss) and the market substitution 100% (54). The paper recycling dataset included the paper production from waste paper minus the avoided virgin production. The benefit of paper recycling was primarily savings in energy consumption, corresponding to a net saving of 1,800 kg CO₂-eq. t⁻¹ paper input (when coal was the marginal energy in the recycling process).

The recycling dataset used for cardboard represented shredding and reprocessing of mixed cardboard and paper materials into cardboard cores and tubes, based on the process operated at *Skjern Paprifabrik*, in Denmark (55). The technical substitution was 100% (55) and the market substitution was assumed 100%, as for paper (54). The cardboard recycling dataset included the cardboard production from waste cardboard minus the avoided virgin production. The benefit of cardboard recycling was primarily savings in energy consumption, corresponding to a net saving of 1,200 kg CO₂-eq t⁻¹ cardboard input (when coal was the marginal energy in the recycling process).

The recycling dataset used for glass represented European glass processes around year 1990, although not necessarily statistically based. It was assumed that cullet technically substituted 99% virgin production (i.e., 1% material loss) by remelting and that the market substitution ratio was 100%. In practice the mixing of cullet and virgin resources for glass production may use a different ratio. The glass recycling dataset included the glass production from cullet minus the avoided virgin production. Any saving in providing virgin resources for glass production was not included. The benefit of glass recycling was primarily savings in energy spending corresponding to a net saving of 260 kg CO₂-eq. t⁻¹ glass input (when coal was the marginal energy in the recycling process).

All inventory data for the recycling processes were based on the EDIP database. Additional information on the GHG accounting for these processes can be found in (56-59).

3.7 Use-on-land

The amount and composition of the compost derived from anaerobic digestion of the source-segregated organic waste and of the bioliquid was calculated based on a mass balance approach, i.e., as the difference between the initial nutrients and dry matter fed to the digestion process and the amount transferred to biogas and lost during the post-composting process. The compost was assumed to be applied on land substituting mineral fertilizers following the approach of (60).

In order to quantify the amount of mineral fertilizers replaced it is necessary to assume a type of soil and a specific legislative context as the application of organic fertilizers is typically regulated differently in each Country. In this study the Danish legislation for application on land of organic fertilizers was assumed as reference for the modelling (61). According to that, the amount of N that can be brought into the field is limited, so that the N cannot be applied in excess. However, not all the N applied translates into avoided mineral fertilizer, as the regulation considered an efficiency of only 20% for compost, i.e. a

substitution rate of 20%. The substitution of P and K was assumed 100% (that is, all P and K applied substituted corresponding amount of mineral fertilizers) conformingly with the approach of (60). This is in accordance with similar studies (e.g. (39)). The distribution of N into NH_3 , NO_3^- , and organic N was set to 13%, 0.2%, and 86.8% conformingly with (60). The air emission of NH_3 and N_2O were assumed to 0.21% (i.e., 1.6% of the NH_4^+ content of the compost) and 1.5%, respectively, of the N applied conformingly with (60, 62). The emission of NO_3^- to water bodies was set to 20% of the applied N according to the findings of (62). It should be noted that N-leaching varied according to soil type, livestock density, and organic waste treatment and was found within a range of 3%-87% of the N added with the waste (61). Based on the results from the same study, the C bound in soil (over the considered 100 years LCA horizon) was assumed 14% of the C initially applied with the compost.

3.8 Collection and transportation

Waste collection was modelled as fuel consumption per tonne of wet waste. The specific fuel consumption has been determined by direct measurements on a large range of Danish collection routes (63). These were ($\text{L t}^{-1} \text{ ww}$): 3 for residual waste, 4 for source-segregated paper, 6 for source-segregated glass, 8 for source-segregated plastic and 6 for source-segregated organic waste.

The fuel consumption for transport was represented by “transportation” to the point of unloading, e.g., at the treatment or disposal facilities. The fuel consumption was expressed in fuel consumption per tonne of waste per km (one-way distance) according to previous studies (63, 64). Transport distances were: 15 km for the residual waste sent to treatment; 40 km for stabilized organic (compost from MBT) sent to landfilling; 25 km for compost from anaerobic digestion of source-segregated organic waste and bioliquid (sent to use-on-land); 70 km for bottom ash (including post-treatment and reuse); 500 km for fly ash (sent to backfilling of old salt mines). As recyclables generally enter a global market, transport distances were unknown in this case but average European values were used for approximation (64): 100 km for plastic, glass and paper, and 500 km for aluminium and ferrous metals.

3.9 Other processes

With respect to incineration residues, bottom ashes from incineration were assumed utilized as construction material substituting natural gravel (36). APC residues were assumed to be utilized in the backfilling of old mines following the approach of (38).

The upgrading of biogas to methane (96% v/v, corresponding to a LHV of 35 MJ Nm⁻³) was modelled based on (44): The electricity use was reported to 0.5 kWh Nm⁻³ CH₄ (0.014 kWh MJ⁻¹ CH₄). The main direct air emissions were CO (1.5E-04 kg CO MJ⁻¹ CH₄), CH₄ (1.5E-05 kg CH₄ MJ⁻¹ CH₄), NO_x (6.7E-06 kg NO_x MJ⁻¹ CH₄), and SO₂ (2.9E-07 kg SO₂ MJ⁻¹ CH₄). Overall, the upgrading process equalled a GHG emission of 0.031 kg CO₂-eq. MJ⁻¹ CH₄ (produced).

4. Waste materials and energy balance

The results of a waste material balance of the assessed waste management scenarios is reported in Table S9. The waste material balance reports the amount of the individual flows of waste material fractions across the investigated scenarios. As reported in Table S9, the amount of source-segregated recyclable materials was the same for all the scenarios assessed (but differed between waste (a) and waste (b)). The amount of source-segregated organic waste varied from zero (scenarios 0), over 70% (scenarios I), to 100% (scenarios II). As a result, the amount of rMSW equalled 690, 430, and 310 kg ww for scenario 0, I, and II for the case of waste composition (a), and 830, 420, 240 kg ww for scenario 0, I, and II for the case of waste composition (b).

Figure S16 illustrates the total amount of electricity and heat recovered in the investigated scenarios. This is further distinguished between bioenergy (from organic, papers, and other biogenic materials) and fossil energy.

Table S9. Waste material fractions balance of the individual scenarios assessed (unit: kg ww). Organic, PA (paper), CB (cardboard), Plastic, Glass, AL (aluminium), and FE (ferrous metal) refer to the source-segregated amount of these waste material fractions; ssMSW: source-segregated MSW; rMSW: residual MSW. Eventual inconsistencies are due to rounding (values rounded to two significant digits).

	Scenarios 0 WR GE, WR TF, INC, MBT AC, MBT DC, BLF, CLF		Scenarios I WR GE, WR TF, INC, MBT AC, MBT DC, BLF, CLF		Scenarios II WR GE, WR TF, INC, MBT AC, MBT DC, BLF, CLF	
	(a)	(b)	(a)	(b)	(a)	(b)
MSW	1,000	1,000	1,000	1,000	1,000	1,000
Organic	0	0	260	410	370	590
PA ^α	180	66	180	66	180	66
CB ^β	38	22	38	22	38	22
Plastic ^γ	8.8	39	8.8	39	8.8	39
Glass ^θ	66	18	66	18	66	18
AL	5.7	3.8	5.7	3.8	5.7	3.8
FE	12	19	12	19	12	19
ssMSW	310	170	570	580	680	760
rMSW	690	830	430	420	320	240

^α Does not include dirty paper which is routed to rMSW. ^β Does not include dirty cardboard which is routed to rMSW. ^γ Does not include non-recyclable plastic which is routed to rMSW. ^θ Does not include non-recyclable glass which is routed to rMSW.

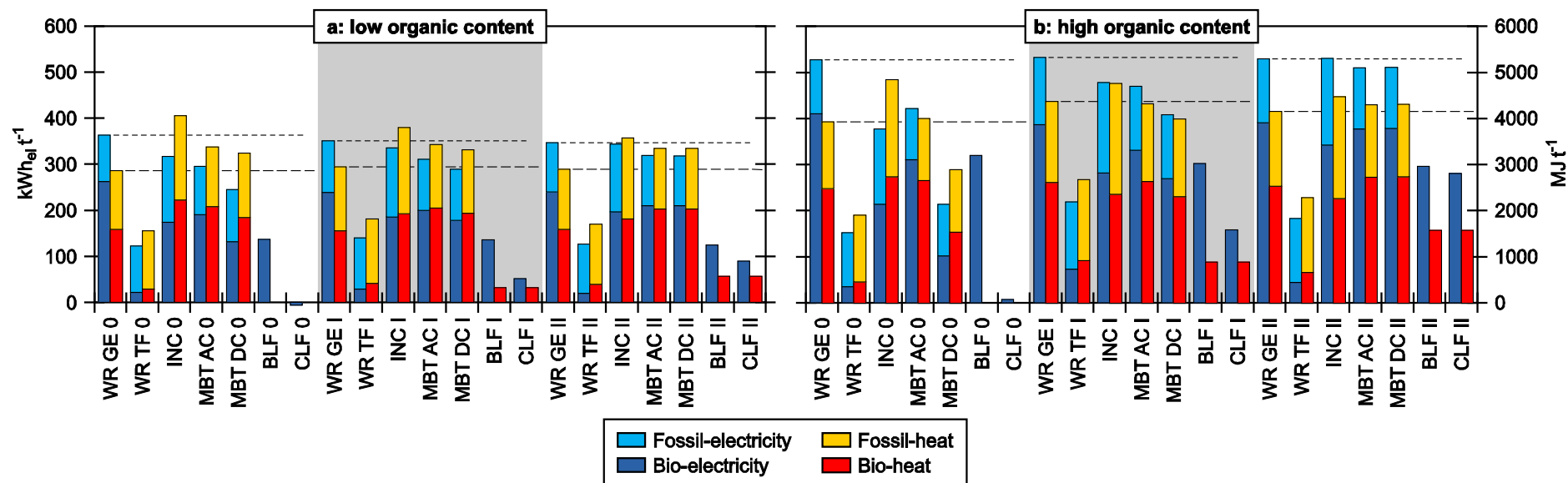


Figure S16. Energy balance: net electricity and heat production in the investigated scenarios. Electricity production refers to the left axis ($0\text{--}600 \text{ kWh}_{\text{el}} \text{t}^{-1}$), while heat production refers to the right axis ($0\text{--}6000 \text{ MJ t}^{-1}$). The energy produced is distinguished between bio- and fossil-based. The dotted lines represent the net electricity and heat produced in the waste refining CHP scenarios (WR GE), here used as comparative reference.

5. Sensitivity analysis: parameter uncertainty (waste refinery vs. incineration)

This section presents the results of a number of sensitivity analyses where the GW performance of two selected scenarios, namely INC 0 and WR GE 0, was compared by individually varying relevant parameters; these were: (i) LHV (i.e., obtained by varying the share of organic waste in the initial MSW), (ii) electricity efficiency of incineration, (iii) electricity efficiency of biogas-to-electricity conversion and (iv) enzymes consumption. The objective was to identify the conditions (with respect to waste composition and technology efficiency) when one waste management scenario allowed for greater GHG savings than the other (i.e., thresholds or break-even points). The results are displayed in Figure S17 for both waste compositions *a* and *b*. The results are expressed as net difference (Δ) between the GW performance of WR GE 0 and INC 0. Therefore, when the indicator is below zero WR GE 0 performs better than INC 0, and viceversa.

With respect to (i) the results were totally dependent upon the selected waste composition: with waste composition *a* incineration was always better than waste refining. This was also true for very low LHV; the reason for this lied in the waste composition as determined by (1): an increase on the share of organic waste determined a decrease of LHV and thus a diminished energy recovery of incineration. However, also the performance of the waste refining scenario suffered as the amount of bioliquid would decrease proportionally to the decrease in the share of paper materials (due to the increased share of organic waste) which dry matter content was significantly higher than that of organic waste. In the case of waste composition *b*, instead, the results were opposite and only for a LHV above 9.5 GJ t⁻¹ MSW incineration performed better than waste refining. In (2), in fact, the measured water content for the organic waste was significantly lower; this benefitted the waste refinery scenario when increasing the share of organic waste (i.e., relatively more dry matter was transferred into the bioliquid compared with the case of *a*).

The result for (ii) again highlighted the importance of the waste composition: in the case of *a* waste refining performed better than incineration only when the electricity efficiency was lower than 15%. In the case of *b*, instead, this occurred always until exceeding 32% efficiency at the incinerator.

Similar results were found for (iii): in the case of *a* waste refining achieved a better performance than incineration only for biogas-to-electricity efficiency greater than 50%. In the case of *b* 36% was instead sufficient.

Lastly, the analysis (iv) revealed that in the case of *a* any decrease in enzymes consumption would not change the ranking between the scenarios. As opposite to this, in the case of *b* waste refining became worse than incineration only for consumptions greater than 15 kg t⁻¹ MSW.

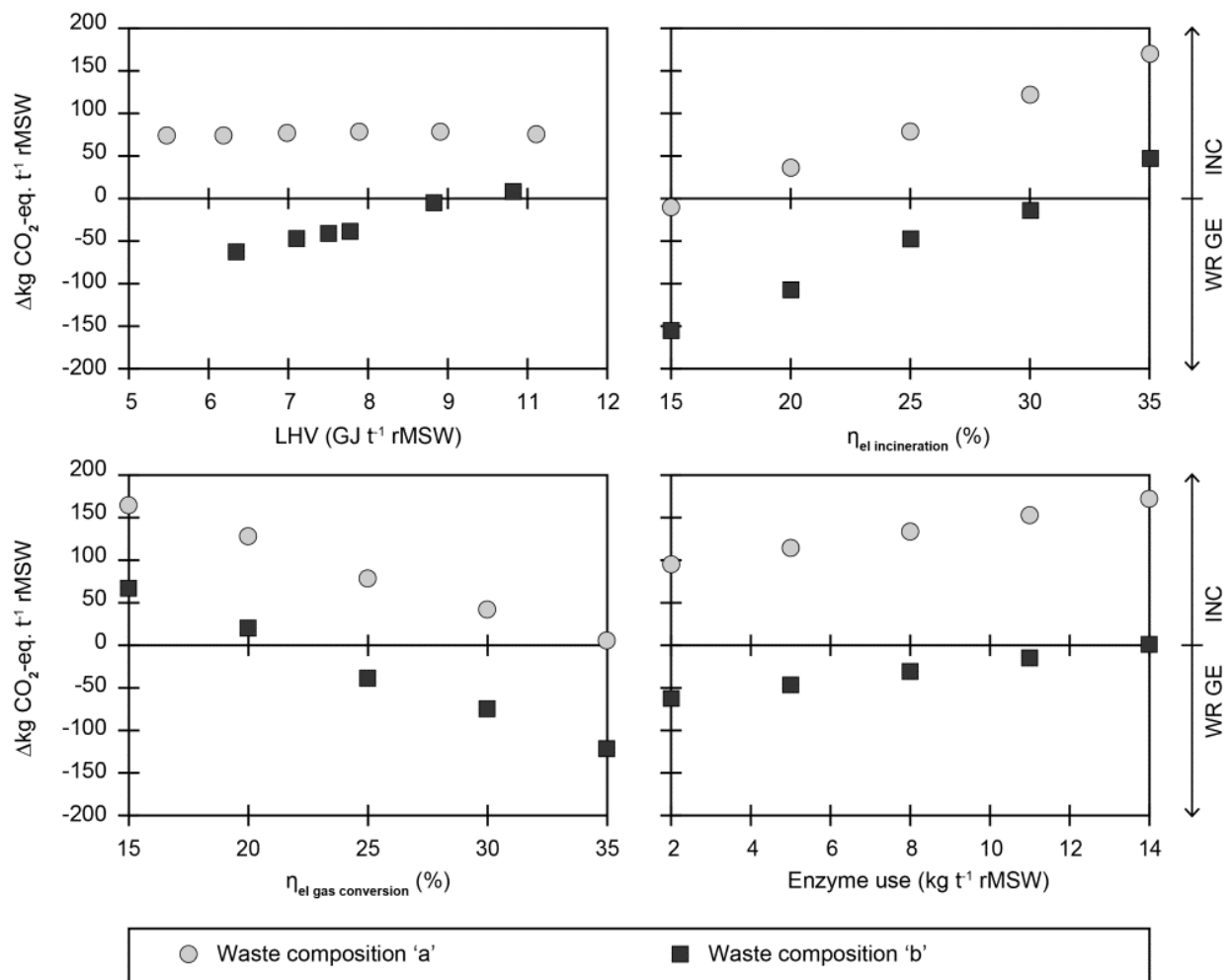


Figure S17. Individual comparison between incineration (INC) and refining with CHP (WR GE) for the scenario without organic waste source-segregation (0). Four selected parameters (LHV, electricity efficiency of incineration, efficiency of biogas-to-electricity conversion, enzyme use) were varied and the corresponding GHG performance of the two scenarios was quantified. The results are expressed as net difference (Δ) between the GW performance of WR GE 0 and INC 0. Therefore, when the indicator is below zero WR GE 0 performs better than INC 0, and viceversa.

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